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- © COLORED UNSATURATED POLYESTER MATERIAL CONTAINING COPOLYMERIZED METHINE DYES, AND PRODUCTS THEREFROM.
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Chemical Abstract, volume 71, no. 12, 22 September 1969, (Columbus, Ohio, US), V.V. Korshak et al.: "Colored polyesters based on bis (hydroxyalkoxy) azo-benzenes", see page 24, abstract no. 50783u, & Izv. Akad. Nauk SSSR, Ser. Khim. 1969, (5), 1078-85

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Description

This invention relates to colored, unsaturated polyesters which contain certain methine dye moieties chemically linked through copolymerization into the polyester backbone whereby the dve moieties become essentially nonextractable from the polyester. These colored polyesters are thus highly suitable for incorporation into curable polyester materials used in the production of bathroom fixtures (sinks, showers, tubs), boats, automotive parts, and the like wherein resistance to die extractability is of great consequence. The addition of the methine die monomers during the polyester preparation also gives greater colorant uniformity than possible, for example, where such colorants are simply blended with the polyester material prior to casting or molding thereof.

It is known, of course, that some methine dyes may be used to color polyester fibers using conventional dyeing procedures. It is also known that certain dyes can be melt blended (not copolymerized) with preformed polyesters or dispersed with reactive solvent (curing monomer) into unsaturated polyester material prior to molding and curing, to produce colored product. In other limited cases, certain dyes such as selected anthraquinones are reactable into the polyester chain by copolymerization.

Advantages of the present methine moieties over others which might be copolymerized into the polyester include (1) greater thermal stability, for example, than azo dyes which generally are not stable to polymer preparation temperatures, (2) methine dyes can be selected or tailored to absorb light at the proper wavelengths to protect particular contents of a package, (3) certain anthraquinone dyes, for example, which absorb light below 420 nm are generally not stable to UV, (4) the present methine dyes have high extinction coefficients, i.e., less dye is needed to give equivalent color, and (5) generally speaking, methine dyes are much less costly than anthraquinone dyes.

In accordance with the present invention, the methine moieties have at least one methine unit defined herein as "the group >C=C< conjoined with a conjugated aromatic system." This unit imparts to the moiety and to the polymer the property of ultraviolet or visible light absorption generally within the range of about 320 nm to about 650 nm. The moieties preferably have molecular weights of from about 200 to about 600 although lower and higher molecular weights are also operable. The moieties are derived from reactants (monomers) having one or more groups which condense during esterification or polycondensation to enter the moiety into the polymer chain. These groups include hydroxyl, carboxyl, carboxylic ester, acid halide, amino and the like. As aforesaid, these methine moieties are thermally stable at polymer processing conditions, for example, including polycondensation temperatures of up to about 300°C. Of course, where only one condensable group is present, the methine monomer would act as a chain terminator in known manner. These moieties are useful in total concentrations ranging from about 1.0 to about 5,000, preferably 2.0 to about 1,500 parts by weight of moiety per million parts by weight of final polymer (ppm).

The present invention is defined in its broad embodiment as unsaturated polyester material having copolymerized therein a total of from 1.0 to about 5,000 ppm, of at least one methine moiety having one or more methine units, said moiety absorbing in the range of from 320 nm to about 650 nm, and being non-extractable from said polyester material and stable under processing conditions therefor.

The extractabilities of the present methine moieties can be determined as follows:

Extraction Procedure

All extractions are done in glass containers with distilled solvents (water, heptane, etc.) under the time and temperature conditions described below. The sample form is 0.5 inch × 2.5 inch (12.7 × 63.5 mm) molded segments of about 10 to 20 ml thickness, the thickness, however, not being critical. All samples are washed with cold solvent to remove surface contaminants and are then contacted with 2 ml of solvent/3.2 cm²) of surface area. After the specified period of ageing in the solvent, the solvent is transferred to glass flasks, concentrated, spiked and analyzed. Solvent blanks (controls) are run under the same conditions without polymer.

Extraction Conditions

1. Water. The samples are added to the solvent at room temperature and heated at 250°F (121°C) (15 psi, 1.03 bar) for two hours. For half the samples, the solvent is transferred to glass flasks, concentrated, and analyzed; the remaining samples are placed in a 120°F (49°C) oven for 30 days and the solvent then transferred to glass flasks, concentrated, and analyzed.

2. 50% Ethanol/water (v/v). The samples are added to solvent at room temperature, placed in an oven at 120°F (49°C), concentrated, and analyzed after 24 hours and 30 days.

3. Heptane. The samples are added to solvent at 150°F (65°C) and heated at 150°F (65°C) for 2 hours. Part of the samples are transferred to glass flasks, concentrated, and analyzed. The remainder of the samples are placed in a 120°F (49°C) oven for 30 days and the solvent then transferred to glass flasks, concentrated, and analyzed.

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Analysis

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instrument — Hewlett-Packard 84504, spectrophotometer

Cell Path Length --- 1 cm

Scanning Range — 400 nm to 650 nm

Calibration for Red Metnine dye in Anisole — 520 nm absorbance maximum

Calibration for Red Methine dye in DMF — 530 nm absorbance maximum

The Hewlett-Packard 8450 A spectrophotometer is equipped with microprocessor and programming capability with RAM memory and floppy disk storage. The instrument is calibrated at 1000 ppb, 1500 ppb, and 2000 ppb dye in anisole and 970 ppb, 1460 ppb, and 1940 ppb in the DMF. The calibration data are stored on a floppy disk for recall when needed.

The extractability of the present methine moieties from the present cured polyester material is essentially nonexistent.

Useful methine reactants or monomers for the present invention have the general formulas:

wherein A is selected from the following radicals designated by their exemplary table numbers:

(10)

wherein:

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R and R' are selected from hydrogen, fluorine, chlorine, bromine, alkyl, alkoxy, phenyl, phenoxy, alkythio, and arylthio; n is 0, 1, 2;

(15)

 R_1 and R_2 are selected from hydrogen; cycloalkyl; cycloalkyl substituted with one or two of alkyl, —OH, alkoxy, halogen, or hydroxy substituted alkyl; phenyl; phenyl substituted with alkyl, alkoxy, halogen, alkanoylamino, carboxy, cyano, or alkoxycarbonyl; straight or branched lower alkenyl; straight or branched alkyl of 1-8 carbons and such alkyl substituted with the following: hydroxy; halogen; cyano; succinimido; hydroxysuccinimido; acyloxysuccinimido; glutarimido; phenylcarbamoyloxy; phthalimido; 4-carboxyphthalimido; phthalimidino; 2-pyrrolidono; cyclohexyl; phenyl; phenyl substituted with alkyl, alkoxy, halogen, hydroxy alkanoylamino; carboxy, cyano, or alkoxycarbonyl; alkylsulfamoyl; vinylsulfonyl; acrylamido; sulfamyl; benzoylsulfonicimido; alkylsulfonamido; phenylsulfonamido; alkoxycarbonylamino; alkylcarbamoyloxy; alkoxycarbonyl; alkoxycarbonyloxy; alkenylcarbonylamino; groups of the formula

wherein Y is -NH-,

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-0-, -S-, or $-0h_20-$; $-S-F_{14}$: $SO_2Ch_2Ch_2SF_{14}$; wherein F_{14} is alky!, phenyl, phenyl substituted with naioger, alkył, alkoxy, alkanoviamino, cyano, or alkoxycarbonył; pyriaył; pyrimidinyi; benzoxazolył; benzimidazovlyl; benzothiazoly:: radicals of the formulae

—OXR₁₆: —NHXR₁₆: —X—R₁₆: —CONR₁₅R₁₆; and —S₂NR₁₅R₁₆; wherein R₁₆ is selected from H, ary! alkvl, and alky substituted with halogen, —OH, phenoxy, ary!, —CN, cycloalky!, alkvisulfony!, alkylthio, alkanoyloxy, or alkoxy; X is —CO—, —COO—, or —SO₂: R₁₆ is selected from alky! and alky! substituted with halogen, hydroxy, phenoxy, aryl, cyano, cycloalkyl, alkylsulfonyl, alkylthic, alkanoyloxy, and alkoxy; and when X is —CO—, R₁₆ also can be hydrogen, amino, alkenyi, alkylamino, dialkylamino, arvlamino, aryl, or furyl; alkoxy, alkoxy substituted with hydroxy, cyano, alkanoyloxy, or alkoxy; phenoxy; phenoxy substituted with one or more of alkyl, carboxy, alkoxy, carbalkoxy, or halogen; R_1 and R_2 can be a single combined group such as pentamethylene, tetramethylene, ethyleneoxyethylene, ethylene sulfonylethylene, or

XR₁₇ ethylene-N-ethylene

which, with the nitrogen to which it is attached, forms a ring; R₁₇ is alkyl, aryl, or cycloalkyl; R₃ is alkylene, arylene, aralkylene, alkyleneoxy, or alkyleneoxyalkylene;

Z is a direct single bond, OCO, O, S, SO_2 , $R_{17}SO_2N=$,

arylene, or alkylene;

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 R_{4} , R_{5} , and R_{6} are each selected from hydrogen and alkyl;

OCNH-arviene

 R_7 is carboxy, carbalkoxy, or $(R)_n$;

R₁₀ is hydrogen, alkyl, and aryl;

 R_{B} and R_{g} are selected from hydrogen and substituted or unsubstituted alkyl, aryl, or cycloalkyl;

 R_{11} and R_{12} are hydrogen, alkyl, hydroxyl, or acyloxy;

B represents the atoms necessary to complete a five or six membered ring and is selected from

$$R_7$$
 R_7
 R_7

Pland Q are selected from evanc, carbalkoxy, carbaryloxy, carbaralkyloxy, carbamyl, carboxy, N-alkyl-carbamyl, N-alkyl-n-arylcarbamyl, N,N-dialkylcarbamyl, N-arylcarbamyl, N-cyclohexylcarbamyl, aryl, 2-benzoxazolyl, 2-benzotniazolyl, 2-benzimidazolyl, 1,3,4-thiadiazol-2-yl, 1,3,4-oxadiazol-2-yl, SC₂ alkyl, SO₂ aryl, and acyl, or Pland Q may be combined as

wherein R_{12} is defined above and R_{18} is CN, COOH, CO₂ alkyl, carbamyl, or N-alkylcarbamyl;

wherein at least one of A, P, and Q for each dye molecule must be or bear a condensable group selected from carboxy, carbalkoxy, carbaryloxy, N-alkylcarbamyloxy, acyloxy, chlorocarbonyl, carbamyloxy, N-lalkyl)₂carbamyloxy, amino, alkylamino, hydroxyl, N-phenylcarbamyloxy, cyclohexanoyloxy, and carbocyclohexyloxy; and

wherein in the above definitions, each alkyl, aryl, or cycloalkyl moiety or portion of a group or radical may be substituted where appropriate with hydroxyl, acyloxy, alkyl, cyano, alkoxycarbonyl, halogen, alkoxy, or aryl, aryloxy, or cycloalkyl. Also in the above definitions, at least one of A, P, and Q for each dye molecule must be or bear a group capable of reacting under polymerization conditions, to incorporate the methine dye into the polymer, including the following, carboxy, carbalkoxy, carbaryloxy, N-alkyl-carbamyloxy, acyloxy, chlorocarbonyl, carbamyloxy, N-alkylcarbamyloxy, amino, alkylamino, hydroxyl, N-phenylcarbamyloxy, cyclohexanoyloxy, and carbocyclohexyloxy, wherein the alkyl and/or aryl groups may contain common substituents such as hydroxyl, cyano, acyloxy, carbalkoxy, phenyl, and halogen which do not interfere with the condensation reaction.

In all of the above definitions the alkyl or alkylene moieties or portions of the various groups contain from 1—8 carbons, straight or branched chain.

The unsaturated polyesters useful in this invention include the esterification and polycondensation products of one or more unsaturated dicarboxylic acids or their anhydrides and one or more dihydric or polyhydric alcohols. As the acid component, the α,β-ethylenically unsaturated polycarboxylic acids include those having 2—12 carbon atoms, e.g., maleic, fumaric, substituted fumaric, citraconic, mesaconic, tetraconic, glutaconic, muconic, and the like, as well as mixtures thereof. Noncurable polycarboxylic acids, i.e., those which do not contain reactive α,β-ethylenic unsaturation, may also be used if employed in combination with one or more of the aforementioned α,β-ethylenically unsaturated polycarboxylic acids. Where such noncurable polycarboxylic acids are employed, the amount thereof should not constitute more than about 80% of the total equivalents of carboxyl groups in the esterification mixture. Preferably, such noncurable polycarboxylic acids will be employed in amounts varying between about 35% and 75% of the above indicated equivalence basis. Among the noncurable polycarboxylic acids that may be used are oxalic, malonic, succinic, glutaric, sebacic, adipic, phthalic, isopthalic, terephthalic, substituted phthalic, such as tetrachlorophthalic, suberic, azelaic, tricarballylic, citric, tartaric, cyclopropanedicarboxylic, cyclohexanedicarboxylic, and the like, as well as mixtures thereof.

As the alcohol component, the saturated aliphatic polyhydric alcohols include those preferably containing only two hydroxyl groups. Among such diols are ethylene glycol, propylene glycol, butylene glycol, diethylene glycol, dipropylene glycol, triethylene glycol, tetraethylene glycol, butanediol-1,2, butanediol-1,3, butanediol-1,4, pentanediol-1,2, pentanediol-1,4, pentanediol-1,5, hexanediol-1,6, neopentyl glycol, 2,2,4-trimethyl-1,3-pentanediol and the like, as well as mixtures thereof. However, saturated aliphatic polyhydric alcohols containing more than two hydroxyl groups may also be employed and include glycerol, trimethylol ethane, trimethylol propane, pentaerythritol, sorbitol, and the like, as well as mixtures thereof. It is usually desirable that such polyols be employed in minor proportions relative to the diol or diols.

The components may be reacted in the manner customarily used in preparing ethylenically unsatruated polyester resins, i.e., at elevated temperatures and atmospheric pressure, although pressures slightly above or below atmospheric may be employed if desired. The reaction temperature is not critical but preferably is just below the boiling point of the most volatile component of the reaction mixture which is generally the alcohol component. However, temperatures in excess of the boiling point of the most volatile constituent may be employed if the reaction vessel has been equipped with a steam-heated reflux condenser which permits water of esterification to escape from the reaction vessel while condensing volatilized reaction components and returning them to the reaction system.

In preparing the curable compositions, the unsaturated polyesters are blended with a reactive curing agent, operably in a weight ratio of agent/polyester of 10/90 to 90/10, but preferably from 30/60 to 70/40. These agents contain one or more CH_2 =C< groups and desirably have boiling points at atmospheric pressure of 60°C or greater. Such agents include styrene, side-chain substituted styrenes such as the amethyl styrene, a-ethyl styrene, and the like, ring substituted styrenes, such as alkyl styrenes, e.g., ortho-

meta and para-alky: styrenes, including o-methy! styrene, p-ethy! styrene, meta-propy! styrene, 2.4-di-methy styrene, 2.5-diethy; styrene, and the like, halostyrenes, e.g., c-bromostyrene, p-eniorostyrene, 2.4-dieniorostyrene, and the like. Also included are alky! esters of acrylic and methacrylic acid, e.g., methyl, ethyl, or buty! acrylate, methyl, methacrylate, and the like. Also usefu are vinyl acetate, vinyl butyrate, vinyl acrylate, acrylamide, acrylamide, methacrylamide and their dderivatives, aliyl compounds such as dially, phthalate, aliyl acrylate, alivl methacrylamide and their dderivatives, alivl c-hydroxyisobutyrate, aliyl trichiorosilane, aliyl acrylate, dially! malonate, dially! davalate, dially! gluconate, dially disconate, dially! sebacate, dially! tartronate, dially! acriate, dially! mesaconate, dially! citraconate, the dially! ester of muconic acid, dially! itaconate, dially! chlorophthalate, dially! dichiorosilane, the dially! ester of endomethyleneterahydrophthalic anhydride, trially! tricarballylate, trially! phosphate, trially! or in combination with one another.

In order to facilitate the curing, it is preferred that a polymerization catalyst be incorporated in the blend at the time of its curing. The type and amounts of these catalytic materials are well known in the art, and any material which normally induces polymerization of polyester resinous compositions can be utilized. The optimum reaction conditions are modified to some extent by the choice of the particular catalyst used in the process. A very active catalyst should be used in lower concentrations, and preferably at lower temperatures, than a less reactive material. The preferred catalysts comprise a wide variety of organic superoxides, i.e., organic peroxides ("acidic peroxides") and hydroperoxides ("alcoholic peroxides"). Mixtures of peroxides and hydroperoxides, including commercially available mixtures such as methyl ethyl ketone peroxide, cyclohexanone peroxide, and the like, are especially effective as catalysts. Among the useful organic peroxide catalysts are acetyl peroxide, benzoyl peroxide, substituted benzoyl peroxides, halogenated benzyl peroxides such as p-bromobenzoyl peroxide, and 2,4-dichlorobenzoyl peroxide, benzoyl acetyl peroxide, phthalyl peroxide, succinyl peroxide, fatty oil acid peroxides, such as coconut oil peroxide, lauryl peroxide, stearyl peroxide, oleyl peroxide, anisoyl peroxide, toluyl peroxide, and the like. Organic peracids, such as peracetic acid and perbenzoic acid, may also be employed. The useful organic hdyroperoxide catalysts include tertiary butyl hydroperoxide, cumene hydroperoxide, diisopropyl benzene hydroperoxide, 1-hydroxycyclohexyl hydroperoxide, the terpene oxides, such as ascaridole, 1-p-methane hydroperoxide, and the like. Various other types of polymerization catalysts may also be employed, for example, compounds such as aluminum chloride, stannic chloride, boron trifluoride, or the azo-type catalysts such as a,a'-azobisisobutyronitrile.

Since the unsaturated polyester may contain a high degree of polymerizable or reactive unsaturation, it is often desirable to blend a polymerization inhibitor therewith to retard internal polymerization of the polyester during any storage period encountered prior to curing. Once the curable composition is contacted with a sufficient amount of a polymerization catalyst, however, the effect of the inhibitor will be overcome. Among the useful inhibitors are phenol, the monoalkyl phenols, such as orthor, meta-, and paracresol as well as mixtures of such isomers, polyalkyl phenols having a plurality of the same or different substituents, e.g., ethyl, propyl, butyl, and higher alkyl radicals attached to their nuclei, catechol, tertiary butyl catechol, hydroquinone, tertiary butyl hydroquinone, resorcinol, eugenol, guaiacol, pyrogallol, benzaldehyde, tannic acid, ascorbic acid, isoascorbic acid, phenylene diamine, sym-di-β-naphthyl-phenylene diamine, aniline, and the like. The amount of polymerization inhibitor employed depends on the nature of the unsaturated polyester as well as the period of storage stability required. Generally, from about 0.001% to 0.3% by weight, based on the total weight of the curable polyester blend will be sufficient.

Inhibitors of this type may be added during preparation of the unsaturated polyester or optionally added later to the curable blend. In addition, other known additives may be employed such as promoters used in conjunction with the catalyst, mold lubricants, fillers and reinforcing materials, other colorants, flow promoters, ultraviolet absorbing agents, and the like.

The conditions necessary for curing the above blends do not depart from the practice ordinarily observed in curing these types of compositions in general. They may be cured in contact with air or in enclosed molds at temperatures ranging from about 10°C to about 160°C, or even higher as long as they are kept below the point at which the particular curable blend employed begins to decompose. Where it is convenient, it is especially desirable to cure the catalyzed blends by heating to between 90°C and about 150°C for a period of about 3 to 90 minutes.

In general, the methine reactants (monomers) are prepared, for example, by reacting the hydrogenated parent of an aromatic moiety A above, which is electron rich, with a Vilsmeier complex to produce an aldehyde [Bull. Societe Chim. de France, No. 10:1898—99 (October 1962); Angewandte Chemie 72, No. 22, 836—845, November 21, 1960]. For reasons of cost and convenience, phosphorus oxychloride (POCl₃) and N,N-dimethylformamide (DMF) are the preferred reagents. Thus, aromatic amines such as anilines, m-toluidines, 2,5-dimethylanilines, 2,5-dimethoxyanilines, or the like are converted in high yields into the corresponding aldehydes via the Vilsmeier reaction, which aldehydes are subsequently reacted with an active methylene compound of the formula P—CH₂—Q in the presence of a base, such as piperidine to produce the methine monomers, as shown in the following reaction sequence

$$F_{0} = \frac{P_{0}}{R_{1}} \xrightarrow{P_{0}} \frac{P_{0}}{P_{0}} \xrightarrow{P_{0}} \frac{P_{0}}{P_$$

Tetrahydroquinolines, benzomorpholines, indoles, thiazoles, and Fischer's base also undergo the Vilsmeier reactions to produce aldehydes. Preparation of the methine monomers and intermediate aldehydes via the above route is disclosed in considerable detail in many patents including: U.S. 2,649,471; U.S. 2,850,520; U.S. 3,247,211; U.S. 3,260,737; U.S. 3,326,960; U.S. 3,349,098; U.S. 3,386,491; U.S. 3,390,168; U.S. 3,453,270; U.S. 3,453,280; U.S. 3,468,619; U.S. 3,504,010; U.S. 3,555,016; U.S. 3,557,434; U.S. 3,652,635; U.S. 3,661,899; U.S. 3,728,374; U.S. 3,787,476; U.S. 3,829,410; U.S. 3,829,461; U.S. 3,846,069; U.S. 3,869,495; U.S. 3,869,498; U.S. 4,879,434; U.S. 3,920,719; and U.S. 4,077,962 It is also known that it is not necessary to isolate the intermediate aldehyde before preparing the methine monomers as the Vilsmeier reaction mixture can be reacted directly to produce the desired product (U.S. Patents 3,917,604 and 4,088,673). All of these patent teachings are incorporated herein by reference.

Since the Vilsmeier complexes also convert hydroxyl groups to halogen, other methods must be employed to prepare methine monomers bearing hydroxyl groups. For example, the monomers can be prepared according to the scheme

comprising reacting N,N-di-β-hydroxyethylaniline with formaldehyde and m-nitrobenzenesulfonic acid in the presence of concentrated HCl and iron filings to produce the intermediate 4'-(di-β-hydroxyethylamino)-benzalaniline-m-sulfonic acid, which can be reacted with active methylenes to produce the monomers (U.S. 2,583,551).

Another method for producing methine monomers containing dicyanovinyl groups is described in U.S. 4,006,178 wherein aromatic amines are reacted with 1-halogeno-2,2-dicyanoethylene to produce corresponding methine compounds as follows:

Intermediate aldehyde compounds containing groups such as acyloxy or alkoxycarbony! can be hydrolyzed to prepare methine monomers containing hydroxy! or carboxy groups, respectively, which are capable of being reacted into the condensation polymer.

The following examples will illustrate the common reaction of an aromatic aldehyde with an active methylene to produce a typical methine monomer.

Example 1

4-(N,N-Dimethylamino)cinnamaldehyde (1,75 g, 0.01 m), methyl cyanoacetate (0.99 g, 0.01 m), methanol (20 mL), and piperidine (3 drops) are mixed and heated together at reflux for 30 minutes. After being allowed to cool, the reaction mixture is filtered. The orange dye (2.5 g) is washed with methanol and air-dried and has the following structure.

30 This yellow dye absorbs light at λ_{max} 464 nm with a molar extinction coefficient of 38,000.

Example 2

Ethyl [[4-(dimethylamino)phenyl]methylene]propenedioate shown below is prepared by the reaction of 4-(dimethylamino)benzaldehyde with diethyl malonate in the presence of a base catalyst in toluene. This pale yellow dye absorbs UV light at λ_{max} 373 nm with a molar extinction coefficient of 33,000.

$$(CH_3)_2N CH=C(COC_2H_5)_2$$

The following tables further exemplify the useful methine reactants.

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Example				
NO.	(R) _D	RI	R2_	P . Q
٠,	==	CH2CH2OH	CH2CH2OH	CN, CN
4	=	CH2CH2OH	CH2CH2OH	CN, SO, CH,
5	3-CH,	CH ₂ CH ₂ OH	CH2CH2OH	CN, CONHCEN
9	3-CH,	CH ₂ CH ₂ OH	CH 2 CH 2 OH	CN, SO, CH3
7	=	CH ₂ CH ₂ OH	CH2CH2OH	CN, COCeHs
8	Ŧ	СН2СН2ОН	CH ₂ CH ₂ OH	CN, ENT
9	3-CH3	си,си(он)си,	C2Hs	CN, CO2CH,
10	2,5-d1-0CH3	CH2CH(OH)CH2OH	Czlls	CN, CONII,
1.1	2-0CH ₃ , 5-CH ₃	C ₂ H ₅	C2Hs	CN, CO, C, H,
13	2-0CH3, 5-C1	C2H5	C2Hs	CN, CO2C6HS
3	2-5CH,	C ₂ H ₅	C2H3	CN, CO2C6H11
14	2-0C, H,	C, H,	C, H,	CN CONNCT

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P . Q	CN, CO2CH1	CN, CN	CN, CO2CH3	CO2C2Hs, CO2C2Hs		CN C. H.	COOH, C, H5	0 CO2 CH3, C6 H5	си, со,си,	СИ, СООН	CN, CO2CH2CH2CH3	CN, CO2CH3	CN, CO2CH1	CN, CN
R2_	C2H5	C ₂ H,OCCH ₃	C, H, OCCH,	с, н, оссн	С2Н,ОССИ3	0 C 2 H 4 O C C H 3	C2Hs	CH 3	си, си, оёси,	сн, сн, осси,	си, си, осси,	CH2CH2OH	CH ₂ C ₆ H ₅	CH2
RI	с, н, оссн,	C2H,OCCH1	с, н, оссн,	е н о с с н з	с, н, оссн	0 C2H,OCCH,	C ₂ H,OCCH ₃	CH 3	C. H.s	C, H s	p-CH3C6H4	m-ClC ₆ H,	CH 2 C & H s	CH2C6Hs
(R) _n	×	Ħ	3-CH ₃	3-CH ₃	ж	Ξ	×	¥	æ	3-CH,	3-0C ₂ H, 0CCH ₃	3-CH,	***	3-CH1
Example No.	15	16	17	18	19	20	21	22	23	24	25	56	27	28

inued)
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'AB

	P, Q	CH3 CN, COC(CH3),	CN, CO,CH;	CN, CO,CH,CH,OCH,	CN, CO ₂ CH,	CH, CO, CH,	CN, CO2CH1	CN, CO,CH,		CN, CN	CN, CONHC, H,-n	CH, CO,CH,	H CN, CN	1 CN, CN	
TABLE 1 (continued)	R2	CH2	CH,CH,OCCH,	CH2CH2OCC6H3	C2Hs	CzHs	CeHs	C2H5	CH2CH2CO2CH1	CH 2CH 2CO 2H	CH2CH2OH	CH2CH2CN	CH 2 COOH	CH2	• •
TABLE 1	R1	CH ₂	CH ₂	CH ₂ , OCH ₃	CH, CH, OCC, H,	CH, CH, OCNHC, H,	CeHs	CH2CH2CN	CH2CH2CO,CH3	CH2CH2CO2H	CH2CH2C6Hs	CH2CH2C1	CH2	CH2	- 11
	(R) _n	3-CH ₃	3-CH3	3-CH ₃	3-CH,	3-CH,	æ	3-CH3	×	Ŧ	æ	H	æ	T	
	Example No.	29	30	31	32	33	34	35	36	37	38	39	40	4.1	

	P, Q	CN, CO,CH,	CN, CO2CH1	CN, CO,CH3	CN, CO2CH,	CN, CO ₂ CH ₃	CN, CO2CH3	CN, CO2CH3	CN, CO2CII3	CN, CO2CH;	CN, CO2CH;	CN, CO, CH,	CN, CO2CH3
		CN,	CN,	CN,	CN,	CN,	CN,	CN,	C _N ,	CN,	CN,	CN,	CN,
TABLE 1 (continued)	R2	-CH,CH,N(SO,CH,)CH,CH,-	OCH1CH1-	-CH,CH,CH,CH,CH,-	-CH ₂ CH ₂ N = 1	-CH2CH2N CCDOH	— СН, СН, N(С, H, 5) SO, СН,	-CH2CH2OC6H5	CH2CH2N CO-CH2	CH2CH2OC2H5	CH2CH2OCH2CH2OC2H5	CH2CH2N COCH-OH	соси-оёси, си,си,
11	RI	-CH 2CH 2N	-CH1CH1-0CH1CH1-	-CH2CH2CH	C ₂ H ₅	C ₂ H ₅	C ₂ H ₅	C2Hs	C ₂ H ₅	CH, CH, OC, H;	CH2CH3	CH2CH1	CH2CH3
	(R) _n	17	3-CH3	Ŧ	3 ~ CH 3	3-сн,	3-CH3	3-CH,	3-CH ₃	×	3-CH3	3-CH ₃	3-CH3
	Example No.	43	44	45	4 6	4.7	48	49	50	51	52	53	40

CO2CH1, CO2CH1

-C2H5

3-CH3

64

CO,CH,, CN

Example No.	(R)	Я	R2_	D' d
		4	COCH	
55	3-CH3	СИз	CH2CH2N CH2CH2	CN, CO2CH1
56	3-CH3	CH2C6H5	CH, CH, N ii	כמי בסיכווי
57	Ħ	CH2C6H11	C2Hs	CN, CO, CH,
58	3-CH,	C2Hs	CH2CH2CH2SO2CH3	CN, CO2CH,
59	3-CH3	C2Hs	CH, CH, OCOC, H,	CN, CO2CH3
09	3-CH,	-CH2CH=CH2	-CH2CH=CH2	CM, CO2CH3
61	3-CH3	-C2Hs	-CH ₂ CH ₂ N CONH	CN, COOH
62	3-CH3	-C2H5	-CH, CH, N CO-CH,	CN, CO,CH(CH
63	3-CH ₃	-C2H5	CH ₂ CH ₂ N	CO2 CH1, CO2C
			CO-CH	

TABLE 1 (continued)

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Example No.	(R) _n	RI	R2	P , Q
65	3-CH3	-C 2 H s	-CH ₁ CH ₂ SO ₂ CH=CH ₂	CO, CH, CM
99	3-CH3	-C,Hs	-CH2CH2SO2CH2CH2SC6H3	CO2CH3, CN
29	3-CH3	-C2H3	-CH ₂ CH ₂ S	CO2CH3, CN
68	3-CH,	-C ₂ H ₅	-CH2CH2SC,H9,	CO2CH3, CN
69	3-CH 3	-C ₂ H ₅	-CH2CH2N(C2H,CN)SO2CH3	CO,CH,, CN
70	3-CH3	-C2Hs	-CH ₂ CH ₂ CONH ₂	CO2CH1, CM
7.1	3-CH ₃	-C2Hs	-CH ₂ CH ₂ CON(C ₂ H ₅) ₂	CO2CH3, CN
72	3-CH3	-C ₂ H ₃	-CH2CH(OCOCH3)CH2OCOCH3 CN, CN	CN, CN
73	3-CH,	-C2H5	-CH,CH,OCC, H,	CN, CH
7.4	3-CH3	-C2 H s	-CH2CH2OCC&H11	CN, CN
75	3-CH1	-C2Hs	-CH2CH2SO2N(C2H5)2	CN, CO2CH3
16	3-CH1	-C2Hs	-CH2CH2SO2N(CH3)C6H5	CM, CO,CH,
7.7	3-CH1	-C2Hs	-CH2CHS2-CNH	CN, CO,CH,
78	3-CH3	-C2Hs	-CH2CH2-S-C2H4OH	CH, CO2CH2CeHs
79	3-CH3	-C2Hs	-CH ₂ CH ₂ S-C _S	CN, CO2CH1

	D d	CN, CO ₂ CH ₃	CN, CU, CH,	CN, CO, CH,	CN, CO, CH,	Z-E	CN, -C C-C, H;	CN, CONH	CN, CO2CH 3	Corchi, CN	CO2CH3, CW	CO, CH, CM
TABLE 1 (continued)	R2	-CH ₂ CH ₂ SO ₂ C ₂ C ₁	-CH, CH, S-C, O	-CH2CH2S-	-CH(CH,)CH,CO,CH,	-CH(CH1)CH2CO2C2H5	-CH2CH2OCH2CH2OH	CH2CH2CCH3C6H3	CH2CH2OCCH3 -CH2CH2OCCH3	-си ₂ си(сеи,)оёси, -си ₂ си(ососи))си ₂ с1	-CH2CH2OCH2C1	-си,си,оёси,оси,
	RI	-C ₂ H ₅	-C, H,	-C2H5	-C2Hs	-C2Hs	-C2H5	-C,H,	-C ₂ H ₅	-C ₂ H ₅ -C ₂ H ₅	-C2H5	-C2H3
	(R) _n	3-сн,	3-CH,	3-CH3	3-CH,	3-CH,	3-CH3	3-CH,	3-CH3	3-CH ₃	3-CH3	3-CH,
Examole	NO.	В 0	81	8.2	93	84	8.5	98	8.7	68 88	0.6	91.

TABLE 2	C = CH	$Q \qquad (R)^{\frac{1}{3}} = 0$
	P	٥ /

Example					
No.	(R) _n	RI	Н3	2	P, C
9.2	=	CzHs	-CH2CH2-	-503-	CM, CO2CH,
93	3-CH,	C,H,	-CH 2CH 2-	203-	CN, CO,CH,
94	3-CII,	CzHs	-CH2CH2-	-0-	CN, CO1CH1
95	3-CH3	C ₂ H ₃	-CH,CH,O-	-CH2CH2-	CN, CO1CH,
96	3-CH,	C,H,	-CH2CH2-	-020-	CN, CO,CH,
16	3-СН,	C ₂ H ₅	-CH2-		CH, CO1CH1
86	3-CH ,	CAHS	-CH 1CH 1-	• I S - S - S - S - S - S - S - S - S - S	CN, CO, CH,
66	3-CH,	C2113	-CH2CH2-	-осси, си, со-	CN, CO,CH,
100	3-CH3	C ₂ H ₅	-CH2CH2-	ос (си ₂), со-	CN, CO1CH1
101	3-сн,	C, H,	-CH 3 CH 3-	-NHC(CH ₂) 6 CNH-	CN, CO, CH,

			TABLE 2 (continued)		
Example No.	(R)n	$\frac{R_1}{}$	R ₃	2	p, 0
102	3-си,	C2H3	-CH2CH2-	-CNHCH3	CN, CO, CU,
103	3-CH ₁	С2Н5	-CH 2 CH 2 -	-02	CM, CO2CH3
104	±	C,1115	-CH 2 CH 2 -	-020-	CH, CO,CH,
105	x	CH, CH, CN	-CH12CH1-	-000-	CM, CO, CM,
106	Ξ	CH1CH1CN	-CH2CH2-	-20-	CH, CO2CH1
107	=	CH ₂ CH ₂ CN	-CH 2 CH 2-	-ос(сн,),со-	си, си
108	×	CH, CH, CN	-CH 2 CH 2 OCH 2 CH 2 -	-02	CN, CN
109	=	CH ₂ CH ₂ CN	-CH 2 CH 2 -	000-00-00	CN, 80, CH,
110	×	CH2CH2CN	-CH ₂ CH ₂ -	-02	CH, 30,C6H3
111	Ŧ	CH, CH, OCCH,	-CH2CH2-	-02	CN, CONHC.

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	2	-02	-81	-оёси, си, си, ёо- о	-оёсн, сн, сн, ёо- о	-05cH2cH2cH2cO-	1	-02	-00	-02	-00	502
TABLE 2 (continued)	R3	-CH2CH2-	-CH 2CH 3-	-CH2CH2-	-CH2CH2-	-CH2CH(CH1)-	-CH2-	-CH ₂ CH ₂ CH ₂ -	-CH2CH2CH2-	-CH2CH2-	-CH2CH1-	-CH 2CH 2-
	R1	C ₂ H ₅	CzHs	C,111,	C.H.s	C, II, 1	CH, CH, CeHs	CH2CH2OC2H3	CH, CH, OC, H;	CH2CH2N COCH2	CH1CH1CH1	CH 2 CH 2 OH
	(R) _n	2,5-dlocH ₃	3-61	2-0CH1, 5-C1	3-CH3	3-сн,	3-CH ₃	3-CH3	3-CH3	3-CH ₃	3-CH,	3-CH,
200	No.	112	113	114	115	116	117	118	119	120	121	122

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	p, 0	CN, CN	CN, CO,CH,	- O - O - O - O - O - O - O - O - O - O		CO2C2H5, CO2C3H5	CO2C2H3, SO2CH3	CN, C.H.	CN, CONB,	COC, H, COC, H,	CO2C4H3, CO2C6H3	CN, CONHC, HLOH	CN, CONIC, II, -n	CN, CONIICAHII	COMH, COMH,
	2	-205-	об (сн., со-	single bond	-0°(CH2),°CO-	-0c(CH ₂), co-	-0°(CH ₂), °C0-	-оё, (сн.,), ёо-	-0;(сн;),со-	-0С(си,),СО-	-од (сиз),со-	-од (си,), до-	-05 (CH ₂), CO-	-0Ё(сн ₂),ёо-	-од (си,), со-
TABLE 2 (continued)	R ₃	.H3 -CH2CH2-	-CH1CH1-	-CH2CH2-	-CH2CH2-	-CH2CH2-	-CH2CH2-	-CH1CH1-	CH(CH,)CH2CO2CH, -CH2CH2-	-CH ₂ CH ₂ -	-CH2CH2-	-CH2CH2-	-CH2CH2-	-CH2CH2-	-CH2CH2-
	RI	CH 2CH 2CO 2CH 3	CH2CH2C1	C2H3	C2H3	C2115	C2H5	C ₂ H _S	Сн (Сн ,) Сн	C2H3	C2Hs	C, Hs	C, H,	C2Hs	C ₂ H ₃
	(R)n	3-CH,	3-CH1	3-CH,	3-CH,	3-CH3	3-CH,	3-CH,	3-CH,	3-CH,	3-CH,	3-CH3	3-CH3	3-CH,	3-CH3
1	No.	123	124	125	126	127	128	129	130	131	132	133	134	135	136

TABLE 3	$=CH - \begin{bmatrix} & & & & & & & & & & & & & & & & & &$

P • Q	CN, CO,CH,	CN, 30, CH,	CO,CH,, CO,CH,	CN, SO1C.Hs	CN, CO2C2H3	CN, CN	CN, CN	CM, CO,CM,	CN, CONHCLIIS	CN, -CO	-0-5-5-70-
R6	CH.	CH,	CH,	CH.	CH,	CH.	CH.)	СН,	CH.	CH.	CII 3
RA, RS	CH, CH,	CH3, CH3	CH, CH,	CH, CH,	CH, CH,	CH1, CH1	CH1, CH1	CH1, CH1	CH3, CH3	CH3, CH3	CH, CH,
R1	C, H,	CH ₂ CH ₂ OH	CH2CH2OH	CH2CH2OH	CH1CH1011	о, и, осси,	C, H, OCNH-	Callicn	C2H, OH	С, Н, ОН	C 2 H 4 OH
(R) _n	×	æ	==	=	7-CH3	7-CH3	7-CH3	7-CH3	7-CH3	7-CH1	7-CH3
Example No.	137	1 38	139	140	141	142	143	144	145	146	147

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	P, Q	-0.000 0 -0.000 0	CN, CONNC, H, OI	COC(CH1)11 CN	CM, CO1CH,	CN, CO1C1H5	CN, CONH,	CN, CO, C&Hs	CN, CO,CH,	CN, CO,CH,	CN, CO2C2H4CN	CN, CO2CH3	CN, CN	CN, 302CH3	CN, CO2CH3	CN, CO ₂ CH ₃
	R6	CHJ	CH,	CH 3	CHJ	CH3	CH &	CH.	CH 3	CH3	CH,	Ŧ	×	=	Ξ	Ξ
(pen	R4 , R5	CH1, CH1	CH, CH,	CH1, CH1	CH, CH,	CH3, CH3	CH3, CH3	CH, CH,	CH), CH,	CH1, CH1	CH3, CH3	H, CH,	H, CH,	H, CH,	H, CH,	н, сн,
TABLE 3 (continued)	$\frac{R_1}{}$	C,H,OH	C ₂ H, OH	C2H, OH	C2H,OH	C ₂ H,OCC ₂ H ₅	C, H, OCC, H,	0 C2H,0COC2H,	CH2C6H3	CH1	CH ₂ C ₆ H ₁₁	C, Hg-n	CH2CH2OCH2CH2OH	CH2CH2OCCH3	CH2CH2N(CeH5)SO2CH3	CH2CH2N II
	(R) _n	7-CH,	7-CH3	7-CH3	=	=	æ	=	32	æ	=	7-CH3	7-CH,	7-CH3	7-CH3	7 -CH2
	Example No.	148	1.49	150	151	152	153	154	155	156	157	158	159	160	161	162

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Example No.	(R)n	RI	R4, R5	R6	0 '4
163	7 - CH 3	CH2CH2N CO-CH2	н, сн,	±	CN, CO1CH1
164	7-CH3	CH ₂ CH ₂ SC S	н, си,	æ	CN, CO2CH3
165	о 5-сн, 8-си, о си,си, осен,	о Сн ³ сн ³ осн ³	B, CH1	Ξ	CH, CO2CH,
166	5,8-di-UCH,	CH ₂ CH ₂ OCH ₃	H, CH,	Ξ	CN, CO,CH,
167	7-0C2H5	CH ₂ CH ₂ OCCH ₃	H, CH,	=	CM, CONHC ₆ H ₁₁
168	7-CH3	CH2CH20CNH-	H, CH,	Ξ	:=•
169	7-CH,	CH 2CH 2CO 2CH 3	H, CH,	Ξ	CN, CH
170	7-CH1	CH2CH2COOH	H, CH,	×	CM, COOH
171	7-CH,	CH2CH2CON(C2H5)2	H, CH,	=	CONHI, COICHI
172	==	CH2CH(OH)CH1	н, н	=	CN, CN
173	=	CH2CH(CeH5)OH	н, н	=	CN, CN
174	7-CH3	CeHs	H, CH,	CH 3	CN, CO1CH1
175	7-CH3	CH2CH2S	н, си,	æ	CN, CO1C1H4OH
176	7-CH,	CH2CH2OC6H3	H, CH,	I	CN, CO,C,H,C]
177	7-CH1	CH2CH2CH2SO2CH2	H, CH,	z	CN, CO2C4H9-n

:		TABLE 3 (continued)	(þe		
Example No.	(R) _n	R1	R4 · R5	R6	P, Q
178	7-Bt	CH ₂ CH ₂ SO ₂ C1	СН3, СН3	CH 3	CO2C2H5, CO2C2H
179	7-01	CH ₂ , COOH	CH3, CH3	CH.	COC&Hs, SO2CH3
180	7-0CH ₃ 7-CH ₃	Си, си (осси,) си, осси, си, си (он) си, он	CH1, CH1	CH.	CN, CN
182	7-CH)	CH 2CH (OH) CH 2OH	CH1, H	æ	0 CONH2 C. H5
183	7-CH3	CH2CH=CH2	CH, H	Ξ	CN, CO2CH3
184	7-CH3	CH 2CH 2N H H L COOH	CH ,, H	Ξ	CN, CO2CH3
185	7-CH3	СН2 СН2 N С СН - ОН	CH1, H	=	CM, CO2CH3

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Example No.	(R)n	R1	R4	0 4
186	=	C2H & OH	Ξ	CN, CN
187	=	C2H4OH	Ξ	CM, SO2CH3
188	=	C ₂ H, OH	I	CN, SO2C6HS
189	Ξ.	C ₂ H ₄ OH	=	CN, CONICE HS
190	×	C2 H, OH	=	CN, CONHC, H, OH
191	=	Си, си, осси,	=	CH, CN
192	z	он, си, осси,	=	си, 30,си,
193	6-CH ₃	си, си, осси,	æ	CN, CO,CH,
194	6-CH3	0 СИ2СИ2ОС-С6И3	3-CH3	CN, CO, CH,
195	6-CH,	си, си, осси,	3-CH ₃	-2-2-20-

	P. Q.	H) -0C-CC+	H3 -0C-C-C-C	H, CN, CO,CH,	H ₁ CN, CN	H, CN, CO,CH,	H ₃ CN, CO ₂ CH ₃	H CN, CN	cm − − − − − − − − − − − − − − − − − − −		H) CN, CO2CH)	н, си, со,сн,	H3 CN, CO2CH3	H, CN, CO,CH,	H1 CN, CO,CH1
	RA	3-CH,	3-CH 1	3-CH 1	J-CH	3-CH,	3-CH 3	3-CH	3-CII,	3-CH ,	3-CH 3	3-CH,	3-011	3-CH,	3-011
TABLE 4 (continued)	R1	CH, CH (OH) CH,	СН, СИ, ОСН, СИ, ОССИ,	CH ₂ C ₆ H ₅	CH ₂ (-CO ₂ CH ₃	Colls	CH2	CH1	CH ₂ CH ₂ OCCH ₃	CH 2 CH 2 CN	CH ₂ CH ₂ N(CH ₃)SO ₂ CH ₃	CH2CH2OCNHC6H3	CH ₂ CH ₂ C1	CH2CH2OC6H5	CH2CH2SC6H5
	(R)n	6-CH ₁	6-CH ₃	6-CH)	6-CH3	6-CH3	6-CH ₂	6-CH3	6-CH ₃	6-CH3	6-CH3	6-CH3	6-CH3	6-CH1	6-CH3
	Example No.	196	197	198	199	200	201	202	203	204	205	206	207	208	209

	P, Q	СИ, СООН	CN, CO2CH3	CN, CO2CH3	CN, CO,CII,	CN, CO2CH3	CN, CN	CN, CO2CH1	CN CO-CH;			CN, CU ₂ CH;	CN, CO2CH3	CN, -C
3	R4	3-CH3	3-CH ,	3-CH,	3-CH3	3-CH3	3-CH3	3-CH3	J-(B)	3-CH3	3-CH3	3-CH 3	3-CH;	3-CH3
TABLE 4 (continued)	RI	CH2CH2S-CH2KH2	CH2 CH2 SO2	CH ₂ CH ₂ N _N CH ₂	-CH2CH1N II	-CH2CH=CH1	-CH2CH2OC2H4OH	-CH2CH2SO2CH=CH2	- CH2 CH (OH) CH2 OH	-CH2CeH11	-CH2CH2SO2N(CH3)2	-CH1CH1CON(CH1)C6H3	0 -CH2CH2OCCH2C1	-CH,CH,OCCH,
	(R)n	6-CH3	6-CH3	6-CH3	6-CH1	6-CH1	6-CH3	6 -CH1	6 - CH 3	6 - CH 1	6-CH3	6-CH,	6-CH,	6-CH3
	Example No.	210	211	212	213	214	21.5	216	217	218	219	220	. 221	222

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	P, 9	CN, -C	CH, -CH, -CH, S	CN, CO ₂ CH ₃	CN, CO2CH3	CN, CO2CH3
(penu	<u>R</u> 4	3-CH3	3-CH3	3-CH,	3-CH,	3 - CH 3
TABLE 4 (continued)	R	о о сиз сиз осси з	д -си, си, осси,	CH,	CH, CH, N C CH,	CH2 CH2 - S-"
	(R)n	6-CH ₁	6 -CH3	6-CH ₁	6 C11 3	6-CH ₁
	No.	223	224	225	226	227

		TABLE 5		
		$\begin{cases} s & s \\ s & l \\ l $	a D = 0	
Example No.	R7	R	R10	P. 1.9
228	æ	CH,	CH,	CN, CO2CH3
229	=	CH,	C. H.s	CN, CU1CH1
230	=	н	C 6 H 5	CN, CO2C2HS
231	=	CH, CH, OCCH,	C ₆ H ₅	CN, 30, CH1
232	Ξ	о Си, си, осси,	S \$ D	CN, SO2C6H5
233	=	он, си, осси,	C.H.s	CN, CN
234	z	ен, си, оёси,	C & H s	ن=، مبر بحص
235	Œ	о Си, Си, ОССИ,	CeHs	CH, COMBCERS
236	=	СИ,СИ,ОИ	Cells	-00-0 -0-0-0-0-

Example		TABLE 5 (continued)	ş	2
No.	<u>R</u> 7	$\frac{R}{1}$	R10	7
237	=	CH 2 CH 2 OH	Calls	CM, CONH,
238	=	CH ₂ CH ₂ OH	CeHs	. V - V - V N D
239	æ	CH2CH2OH	CiHs	CN, CO2C6HS
240	z	CH ₂ CH ₂ OH	Cells	CN, CONH,
24.1	æ	CH,	C & H s	-0
242	=	CH. 3		CN, CO, CH,
243	=	СИ,	18	CN, CO ₂ CH ₃
244	Ŧ	CH,		CN, CO2C2H4OH
245	5-CH3	си,	CH 3	CN, CO2CH,
246	5-01	CH,	CH3	CN, CO, CH,
247	5-0CH,	CHJ	CH3	CN, CO, CH3
248	=	CH2C6H5	C e H s	CN, CO2CH3
249	æ	CH ₂	CaHs	CN, CN
250	×	CH1	C. e. II s	CN, CN
251 252	z =	CH ₂ CH ₂ CN CH ₂ CONH ₂	C ₆ H ₅	CN, CO ₂ CH ₃ CH ₂ C

	P. Q	CN, CO, CH,	CN, CU,CH,	CN, CO, CH,	CN, CO,CH,	CN, CO2CH3	CO ₂ CH ₃ , CO ₂	CN, COOH	CN, CO, CH, C. H,	CN, CO2CH2C6H11	CN, CO, CH,	CN, CO, CH,	CH, CO2CH1	CH, CO, CH,
t .	R10	C&Hs	Calls	Cells	C, H, S	==	×	=	Cells	C & 11 , -p-C0011	C.HP-CO2CH1	CeHs	CeHs	C & H s
TABLE 5 (continued)	$\frac{R_1}{c}$	СИ2СИ2СИ2ИНСС6И5	CH, CH, CH, NHSO, C, H,	CH2CH2CH2N(CH3)SO2CH3	CH2CH2CH2N(C2H,OH)SO2CH1	CH ₃	СН,	CHJ	CH,	CH3	C, 119 - n	H ₂ N	CH, CH, CH, N COCH,	CH2CH2CH2N C-CH2
	R7	Z	=	æ	=	I	=	Ξ	Ξ	=	=	=	æ	z
	No.	253	254	255	256	257	258	250	260	261	262	263	264	265

TABLE 5 (continued)

P. Q	CN, CO, CH,	си, со,сн,
<u>R</u> 10	CeHs	C&Hs C&Hs
<u>R1</u> 9	CH2CH2CH2N	CH2CH2CH2N
<u>R</u> 7	=	ж ж
Example No.	266	267

	TABLE 6	9	
	P C=HC	S R R B	
RB	<u>R</u> 9	$\overline{R}10$	P-19
CH,	CH3	CH,	CN, CO2CH3
CH,	CH,	CHJ	CM, CO1C1Hs
CH,	CH.)	Calls	CN, CO2CH3
CH,	CII.3	C&HS	CN, CUUH
CH3	CH3	Cells	CO1C1Hs, CO1C1Hs
CH 3	CH,	C.B.S	CM, CONIC, II, OH
CH.	CHICHION	C&HS	CN, CN
CH3	CH2CH2OH	CkHs	CN, SO1CH1
CH3	CH2CH1OH	C.B.	CN, SO2C6HS
CH,	CH2CH1OH	Cells	CN, CONICEUS
CH3	CH2CH2OH	Cells	ر مار کرد د د د د د د د د د د د د د د د د د د د
CH,	CH 2 CH 2 OH	CeHs	0 0 CN Ç.H.5 -0C-C=C-

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	P. Q	CN, CN	CM, CM	CN, CO,CH,	CN, CO2CH2CH2CH	CN, CO ₂ CH ₃	си, со ₂ си ₃	CN, CO ₂ CH ₃	CN, CN	CN, CO2CH3	-CN, -C	-CH, -CH, -CH, -CH, -CH, -CH, -CH, -CH,
inued)	R10	C6H5	CeHs	Cills	CeHs	CeHs	C ₆ H ₅	C. 6 H s	C.B. IS	æ	C eH s	C & H s
TABLE 6 (continued)	<u>R</u> 9	CH ₂ CH ₂ OCCH ₃	CH,CH,OCOC,H;	C2115	C6Hs	· · · · · · · · · · · · · · · · · · ·				CH,		
	<u>R</u> 8	CH)	CII,	C2Hs	C2H5	C2Hs	C ₂ H ₅	C ₂ H _S	CH2CH2OH	CH ₃	CH2 CH2 OH	СИ2СИ2ОН
Ехатрје	No.	281	282	283	284	285	286	287	288	289	290	291

TABLE 6 (continued)

P. Q.	CN, - S - NO	CN, SO ₂ (1)	CN, SO ₂ C1	CN, SO ₂ (CN, CO2CH,	CN, CO,CH,	CN, CO2CH3	CN, CO2CH3	CN, CO ₂ CH ₃	CN, CO ₂ CH ₃	CN, CO2CH3	CN, CO ₂ CH ₃
R10	Cells	C & 11 s	C & H s	CeHs	CEHS	Cells	CiHs	C 6 11 5	5	5		Br
<u>R</u> 9	, u	1 11	11		CH 2 C 8 H 5	CH 2 Ce H s	Cellii	C. II9-n	CH ₃	C.B.S	C ₆ H ₅	C. H. s
R8	CH ₂ CH ₂ OH	CH1CH1OH	CH, CH, OH	CH2 CH2 OH	CH.	CH1 C6 H3	CH3	C, 119 - 11	CH,	Сн,	CH3	CH,
Example No.	292	293	294	295	296	297	298	299	300	301	302	303

5
Continued
_
<u> </u>
TAR

<u>Р. д.</u> 9. соон с _е н _s	-0ë-è	3-20-
R10	·s	5
$\overline{R}9$	CeHs	C ₆ H ₅
R ₉	CH 3	C, H, - n
Example No.	304	305

		P 1 9	CN, CO2CH,	CN, CO2CH2CH2CN	CM, CO2CH2CH2OH	CN, CO2CH2CH2C6H5	CN, CO2CH2C6H11	CO1C1HS, CO1C1HS	COOH, CN	-0	CN, CO2CH,	CH, CO,CH1	CN, CO2CH3	CN, CO,CH,	CH, CH
2.7	$\begin{pmatrix} \mathbf{c} & \mathbf{p} \\ \mathbf{c} & \mathbf{c} \\ \mathbf{H} & \mathbf{n} \end{pmatrix}$	R12	=	×	Ŧ	H	Ŧ	=	x	Ŧ	=	=	æ	×	=
TABLE 7	R ₁₂ 2 3 - 1 2 3 - 1 2 3 - 1 3 3 - 1 3 3 3 - 1 3 3 3 3 3 3 3 3	$\frac{R}{11}$	×	=	æ	Œ	2	=	я	::	=	==	±	=	Ю
		(R)n	==	=	æ	=	Ŧ	=	×	¥	5-CH3	5-0CH3	5-01	5,7-di-CH1	5-CH3
		Example No.	306	307	308	309	310	311	312	313	314	315	316	317	318

	P. 9		CH, -K	CH,	CN, SO ₂	CN, SO ₂ (C1	Ĭ	5 : \$ - 2 = 2 - 20 - 10 - 10 - 10 - 10 - 10 - 10 -	CN, CN	CN, CO2CH3	CN, CO2CH3
	R12	Ξ	=	Ξ	Ξ	=	æ	Ξ	I	=	=
TABLE 7 (continued)	R11	но	HO	HO	Ю	ОН	НО	HO	00002H3	0 0 0 0 0	oecu,c1
	(R)n	5-CH,	5-CH3	5-CH,	5-CH3	5-CH3	5-CH3	5-CH1	5-CH3	5-CH1	5-CH1
	Example No.	319	320	321	322	323	324	325	326	327	328

	P 2 9	CN, CN	CN, CN	CH, CO2CH,	CN, CO2CH1	CN, CO2CH1	CN, - K	CN, - 1 CH,	CN, - 1 - 0CH,	CN, - V	CN, CN	CN, CO2CH1	CN,,NO,	CH, CO, CH,
	R12	10	OCCH,	Ξ	CH.)	Ξ	Ξ	=	×	Ξ	=	æ	Ξ	×
TABLE 7 (continued)	R11	HO	о ^д сн.	CII,	CH.	OCNIC ₆ H ₅	ogen, och,	9сн, ос. и,	ойси 2 С « и з	осинстия	06-50	000-00	, 0 0 0 0 0	о осси ,
	(R)n	×	Ŧ	x	ж	Ξ	=	=	=	Ξ	Ŧ	æ	Ξ	=
	Example No.	329	330	331	332	333	334	335	336	337	338	339	340	34.1

	TABLE 8	
	S CH 3 CH 3 CH - CH = C P C P CH = C P C P C P C P C P C P C P C P C P C	
Example No.	<u>R</u> 7	P. Q
342	=	CN, CO,CH,
1	±	CO2CH2CH3, CO2CH2CH3
344	==	CO,CH, SO,CH,
345	=	CO2CH3, SO2C6H3
346	=	CN, CONIC, H, OH
347	Ŧ	CN, CO2CH2CH2OH
348	×	CN, CO2CH2CH2OC2H5
349	×	CN, CO2CH2CH2C1
350	Ŧ	CN, CO2CH2CH2C6H5
351	I	CN, CO2CH2CH2OC6H5
352	=	CN, CO2C6HS
353	Ξ	CN, CO2C6H11
354	=	CN, CO2CH2C6H11
355	æ	CN, CO2CH2C6H3
356	5-CH ₃	CN, CO2CH2CH(CH3)2
,		

	P. Q	CN, CO2CH2CH2CR	CN, CO2CH2	CN, CN	CN, CO2CH3	CM, COOH	CN, CONECEII,	CN, CONIC, II,	CN, SO, CH,	CN, SO, Calls	CN, COC(CH))	o: U		0 CN C.H; -0C-C=C- 0 CO,CH; C.H;	-0 ² -¢ <u> </u>	No de la companya de	CH, TO
TABLE 8 (continued)	R ₇	5-0CH ₃	5-C1	5-соон	5-0001	5-C00II	5-C00H	5-COOH	5-C00H	5-C00H	5-CO ₂ CH ₃		5-CO,CH,	5-CO ₁ CH ₃	5-CO ₂ CH 3	5-CO ₂ CH ₃	5-00,011
	Example No.	357	358	250	360	361	362	363	364	365	366		367	368	369	370	37.1

	P. Q C1	CN, SO ₁	CN, CO2C2H;	CN, CO,CH,	CN, CO,CII,	CM, CM	CN, CO2CH3	CN, CO, CH,	CN, COOH			CO2CH3, -	CN, -C	CN, COMH
TABLE 8 (continued)	R7	5-C0 ₂ CH ₃	5-CO2CH2CH3	5-CO2CH2C6H3	5-CO2CH2CH2OH	5-CO2CH2CH1C6H5	5-CO2CH2CH2CN	5-CO ₂ CH(CH ₃) ₂	5-C00H	5-СООН	8-соон	1000~S	5-C00H	5-C00H
	Example No.	372	373	374	375	376	377	378	379	380	381	382	383	38 4

		0 74	CN, CO2C2H5	CN, CO,CH,	CN, CO, CH,	CH, CO, CH, CH	CH. CO.CH.CH, OCH.		CN. CO, CH, CH, CR, Ce H,	CN, CO, CH, CH, OC, US	CN, CORRES, N, OH	CN. CO, CH.	CHO COO CHO	CN, CO, CN,	CO, C, Hs, CO2 C2 Hs	CN, CO, CH,	• • • • • • • • • • • • • • • • • • • •
	Q O O O O O O O	$\frac{R_2}{R_2}$	CH.	CH,	CII.	C ED :	CII)	C 2 H s	C ₂ H ₅	C 2 11 5	U. 6H9 - II	C2Hs	C 2 H 5	C 2 H 5	Cens	C.6.115	C6 H &
TABLE 9	R ₂ (R) (R)	R	CH ₃	CH3	CH3	CH3	CH.	C2Hs	C2H5	C2H5	C. H. P. n	CH, CH (CH,),	CbHs	Cells	C6115	Colls	CeHii
		(8)	1737 H	=	3-CH3	3-00,113	3-01	2-0CH3, 5-CH3	2,5-d1-OCH,	2,5-d1-CH,	3-CH3	3-C, H,	=	3-0CH3	æ	=	=
		Example	NO.	386	387	388	389	390	391	392	393	394	395	396	397	398	999

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	P, Q	CN, CO ₂ CH ₃	CN, CN	CN, CN	CN, SO ₂ CH ₃	CN, -CON C.H.	-0c-c	CN, CO ₂ CH ₃	CN, CN	مين کيد	0=0
ued)	R2	CH ₂	CH 2 COOH	CH2	CH2		CH 2	CH2CH2C1 C2H4	CH 2 CH 2 OH	CH2CH2OH	ен,си,осси,
TABLE 9 (continued)	R1	CH2	CH ₂ COOH	CH1	CH2	CH1	CH 2 C1	CH2CH2CI	CH ₂ CH ₂ OC ₂ H ₃ CC ₂ H ₃ CC ₄ H ₃ CC	CH2CH2OH	си, си, осси,
	(R)n	::	×	¥	Ŧ	Ŧ	3-CH3	3-CH ₃	3-CH ₃	3-CH ₃	3-CH3
	Example No.	400	401	402	403	404	405	406	407	409	410

	D10	CN, -	CN, CN CN, SO ₂ CH ₃	CN, SO2C6H5 CN, SO2C6H11	CN, CN	CN, CN	CN, CO2CH3 C1	CN, SO ₂ C1	CN, -CN, -CN, -CN, -CN, -CN, -CN, -CN, -	си, со,си,	CN, CO, CH1	
(per	<u>R</u> 2	ch,ch,occh,	C 2 H 5 C 2 H 5		CH,CH,OCOC,H;	CH,CH,OČOC,H;	CH2CH2OCHH-	си, си, оёс, и, ,	CH,CH,OCC,H,	Си 2 Си 2 ОС - 0	CH2CH2OCCH2	
TABLE 9 (continued)	R	си, оёси,	CH,CH(OH)CH,OH	си,си(оёси,)си,оёси, си,си(ой)си,	сизсизососяня	CH, CH, OCC, H,	CH, CH, OCNH-	CH2CH2OCC2H5	CH, CH, OCC, 115	си, си, оё-	CH2CH2OCCH2	
	(R)	3-CH,	3-CH ₃	3-CH3	3-CH3	3-CH3	3 - CH3	3-CH,	3-CH3	3-CH3	3-CH ₁ 3-CH ₁	
	Example No.	411	412	414	416	417	418	419	420	421	422	

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$$C = HC = \begin{cases} R_{1} & R_{2} \\ Q & \\ Q & \\ R & \\ R$$

P. L. Q	CN, CO2CH3	CN, CO2C2H,OH	CN, CONIC, II, OH	CN, CO2CH2CH2CH	CN, CO2CH2CH2OC3H5	CN, CO2CH2CH2C6H5	CN, CO2CH2C4H3	CN, CO2CH2CH2OC6H5	CN, CO2C2H5	CO2C2Hs, CO2C2Hs	CO2CH3, SO2CH3	CN, CN	CN, SO2C4H5
9	CHJ	CH,	CH.	CH.	CH.	CH.	CH3	CH.	CH 1	CH,	CH 3	CH,	, CH,
R4 L R5 R6	CH, CH,	CH1, CH1	CH1, CH1	CH1, CH1	CH1, CH1	CH3, CH3	CH3, CH3	CH1, CH1	CH3, CH1	CHJ, CHJ	CH, CH,	CH1, CH1	CH3, CH3
$\frac{R}{1}$	CH,	CH,	CH,	CH,	CH1	CII 3	CH,	CH,	CH2CH2C1	CH2CH2OH	CH2CH2OC.H5	о си,си,оёсн,	си, си, оёси,
(R)n	Ŧ	×	æ	×	¥	=	Ħ	æ	Ŧ	=	æ	Ξ	=
Example No.	424	425	426	427	428	429	430	431	432	433	434	435	436

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	P. L.	CN, SO,CH,	CN, CONHC&HS	CH, CONHC, H, OH	CN, SO2CH,	CN, CO2CH,	مين مين مين مين	CN, CONH	0 CO CO CH C C H S		- 0===	CN, CO2CH3	CN, CO2CH3
	<u>R</u> 6	СН	CH.	CHJ	I	C&HS	CHJ	CH 3	CH.	CH.	CH.	CH,	CH 3
ontinued)	R41_R5	CH3, CH3	CH), CH)	CH1, H	н, н	H , H	H . H	н, н	н, н	н, н	= '= =	# ' # .	н 'н
TABLE 10 (continued)	R1	си, си, оёси,	cu, cu, occu,	ch, ch, occu,	сн, сн, оёсн,	CH, CH, OCCH,	сн, сн, оёсн,	сн ₂ сн ₂ оссн ₃	CH 2 CH 2 OCOC 2 H 5	CH, CH, OCC 6H5	CH2CH2OCNH-	CH2CH2CH2NHC-	CH2CH2C1
	(R)n	=	×	z	Ŧ	=	6-C1	7-CH,	×	æ	×	Ξ	=
Example	No.	437	438	439	440	441	442	443	444	445	446	447	448

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	P. L.	CH, CO,C.H;	CN, CO2CH2C6H5	CN, CN	CN, CN ,^C1	CN, CN,	CN, LL, NO	CN,	CN, SO ₁	CN, CO2CH2CH2CH
	<u>R</u> 6	CH.	CH.	CH 3	CH.	CH 3	CH.	CH 3	CH,	CH 3
inued)	R41. R5	H , H	# ' #	H , H	н, н	H .	H .	н , н	н н	H , H
TABLE 10 (continued)	RI	CH, CH, CH, N COCH,	CH2	CH2	CH2	CH2	CH2	CH2	C4H5	0 CH2CH2OCCH3C6H3
	(R).n	=	æ	=	=	=	z	Ξ	= =	=
	Example No.	449	450	451	452	453	454	455	456	458

	P. C. 2	CN, COC(CH,);	CN, COC. Hs	си, соон	CN, C6Hs	CN, S	си,
	R_6	CH,	CHJ	CH3	CH3	CH 3	CH.
TABLE 10 (continued)	R41_R5	н , н	H .	н, н	н, н	ш 'н	H .H
TAB	R1 0 CH2 CH2 OCCH3		си, си, осси,	CH2CH2OCCH3	CH, CH, OCCH,	CH2CH1OCCH1	си, си, осси,
	(R)n		I	Ŧ	=	¥	z
	Example No.	459	460	461	462	463	464

		P. 1. Q	CN, CN	CN, CONII,	CN, CO2CH1	CN, CONICEUS	0::0		CN, CN	CN, CO2CH3	CN, SO2CH3	CN, SO2CeHs
TABLE 11	R_{1} $-H_{-1}$ $-H_{-1$	<u>R1</u>	CH 2CH 2OH	CH2CH20H	CH 2 CH 2 OH	CH 2 CH 2 OH		CH ₂ CH ₂ OH	ch20ch2	CH ₂ CH ₂ OCCH ₃	CH ₂ CH ₂ OCCH ₁	CH, CH, OCCH,
Ħ	$C = HC - \frac{1}{4}$ Q (R)	(R')n	Ħ	Ŧ	Ŧ	Ŧ	-	Ξ	æ	Ħ	×	Ξ
		(R)n		: I	: =	×		=	Ξ	=	æ	Ξ
		Example No.	465	466	467	468		469	470	471	472	473

Eramole		TABLE	TABLE 11 (continued)	
NO.	(R)n	(R'In	$\frac{R_1}{L}$	Pr. Q. 19
474	×	=	си, си, осси,	CN, -C .N.O
475	×	Ξ	сн 20 с с 1 2 с с 1 3	-0=-00-
476	×	æ	CH ₂ CH ₂ OCCH ₃	0 CN C6H3 -C-C-C-
477	Œ	Ŧ	си,си,овси,	CN, COC. Hs
478	=	×	CH ₂ CH ₂ OCCH ₃	CN, COC(CH;),
479	=	×	0 CH,2CU,2CU,2H,3	CN, CeHs
480	=	н	CH2CH2C1	CN, CO2CH3
481	3-CH1	Ŧ	CH2CH2OH	CN, CO, CH,
482	3-CH3	•	C ₂ H ₅	CN, CO,CH,
483	3-CH3	Ħ	CH ₂	CN, CO,CH,
484	=	I	CH 3	CO2C1H1, CO1C1H5
485	=	æ	CH2CH(OH)CH3	-0
486	æ	ж	CH ₂ COOH	CN, CN
487	×	Ξ	CH 2 CO 2 CH 3	CN, CN

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3-CH ₃ 3-CH ₃ 3-CH ₃ 3-CH ₃ 3-CH ₃ CH ₂ 2,5-d1-OCH ₃ H CH ₃ CH ₂ 2,5-d1-CH ₃ H CH ₃ H CH ₃ H CH ₂ CH ₃ H H CH ₂ CH H H CH ₂ CH H H H CH ₂ CH CH ₂ CH H H H CH ₂ CH CH ₂ CH H H H CH ₂ CH CH ₂ CH H H H CH ₂ CH CH ₂ CH H H H CH ₂ CH CH CH CH ₂ CH CH CH CH CH ₂ CH C	Example No.	(<u>R)</u> n	TABLE TABLE	TABLE 11 (continued) $\frac{R_1}{CH_2 - CO_2 CH_3}$	P. Q.
3-C1 ₃ 2'-CH ₃ CH ₂	• • • • • • • • • • • • • • • • • • •	3-CH,	3'-CH ₃	CH ₂	CN, CO1CH1CH
3-CH ₃ 3'-OCH ₃ CH ₂	490	3-c1,	2'-CH3	CH ₂ *CO ₂ CH ₃	CH, CO2CH2CH2OC2H3
2,5-d1-OCH ₃ H CH ₃ 2,5-d1-CH ₃ H CH ₃ H CH ₃ H CH ₃ H CH ₄ C ₂ H ₅ H CH ₂ CH ₃ H CH ₂ CH ₃ H CH ₂ CH ₃ C ₆ H ₅ H CH ₂ CH ₂ CO ₆ H ₅ H CH ₂ CH ₂ CO ₆ H ₅ H CH ₂ CH ₂ CO ₆ H ₅ H CH ₂ CH ₂ CO ₆ H ₅ H CH ₂ CH ₂ CO ₆ H ₅	491	3-CH,	3'-0CH3	CH1	CN, CN
2,5-di-CH, H CH, 3-Br 3'-CH, CH, H R CH2CH(CH,); H H CH2CH(SH); H H CH2CH2CN H H CH2CH2SO2CeH5 H H CH2CH2CN	402	2,5-d1-0CH,	Ŧ	CH ₂	CN, SO ₂ CH ₁
3-Br H H CL2H5 H H CH2CH(CH1)1 H CH2CH2SO2C6H5 H H CH2CH2SO2C6H5 H H CH2CH2SO2C6H5 H H CH2CH2SO2C6H5 H H CH2CH2SO2C6H5	, c.	2,5-d1-CH,	**	CH ₃	CN, CO ₂ CII ₃
H H CH ₂ CH(CH ₃) ₂ H H CH ₂ CH(CH ₃) ₂ H CH ₂ CH ₂ SO ₂ C ₆ H ₅ H H CH ₂ CH ₂ SO ₂ C ₆ H ₅ H CH ₂ CH ₂ SO ₂ C ₆ H ₅ H CH ₂ CH ₂ SO ₂ C ₆ H ₅	494	3-8r	3'-CH3	CH,	CN, CO2CH3
H CH2C6H5 H CH2C6H5 H H CH2C6H5 H H CH2CH2CN	495	- E	×	C2Hs	CN, CO2CH3
H CH2C6H5 H B CH2CH2SO2C6H5 H B CH2CH2CN H B CH2CH2CN H B CH2CH2C6H5 H B CH2CH2S-E	496		Œ	CH ₂ CH(CH ₃) ₂	CN, CO2CH3
H CH2CH2SO2C6H5 H CH2CH2CN H H CH2CH2CN H H CH2CH2OC6H5 H H H CH2CH2S-C () () () () () () () () () (497	: =	7	CH2C6H5	CN, CO2CH3
H H CH2CH2CN H H CH2CH2OC&H5 H H CH2CH2S-E	4 Q B	: =	. 	CH2CH2SO2C6H5	CN, CO2CH3
H CH2CH2OC6H5 H CH2CH2S-C, , , , , , , , , , , , , , , , , , ,	064	: =	=	CIIZCHZCN	CN, CO2CH1
И СН2СИ2S-С	500	: ==	x	CH2CH2OC6H5	CN, CO2CH1
	, US	: ==	æ	CH2CH2S-E	CH, CO2CH1

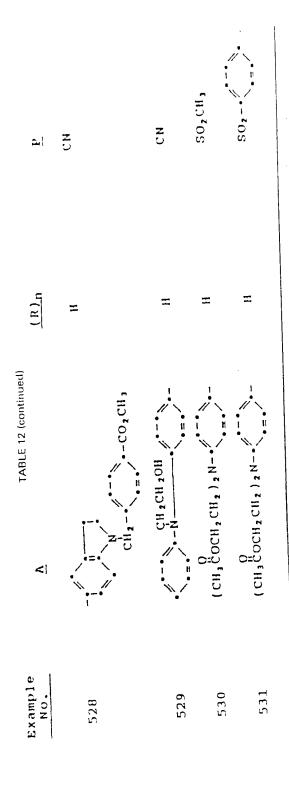
	P. Q	CN, CO ₂ CH ₃	CN, CO2CH1	CN, CO ₂ CH ₃	CN, CO ₂ CH ₃	-0C-C	CN, CO1CH1CH2OH	си, соинстикон	
1 (continued)	$\frac{R}{\Box}$	CH ₂ CH ₂ N ₄ H ₂ CH ₂	CH ₂ CH ₂ -N H H SO ₂	CH ₂ CH ₂ N SO ₂ - H	CH 2 CH 2 SO 3 NH	CH 2	CH1	CH ₂	
TABLE 11 (continued)	(R')n	Ξ	=	=	Ξ	Ξ	Ŧ	=	
	(R)n	=	×	Ŧ	=	=	=	==	
	Example No.	502	503	504	505	905	507	508	

TABLE 12	$A-CH=C-\begin{pmatrix} P & & & & & P \\ & & & & & & \\ & & & & &$

<u>ಒ</u> !	CN	CN	CN	CO2CH3	CO2CH3	CO3 CH3	CONIL,	
(R)n	x	Ξ	æ	Ξ	±	2-CH,	2-C1	
< I	(HOC ₂ H ₄) ₂ N	NN	CH, EOCH, CH, N	(CH ₃) ₂ N-•(C ₆ H ₅ CH ₂	Ce H11 N- N- C2 H5	CH ₃ COOCH ₂ CH ₂ CH ₃ COOCH ₂ CH ₂	
Example No.	509	510	51.1	512	513	514	515	

	ᆈ	CN	Z U	C	N	m O	CN
	(R)n	Ξ	2,5-di-CH,	Ξ.	=	æ.	2-0CH ₃
TABLE 12 (continued)	A CH 3		CH1 CH1) 2 CH2) 2	си, осси, осси,	CH ₃ -1 CH ₃ CH ₃ CH ₃	N C & H S C C B L C C B L C C C C C C C C C C C C	S C H S O C C H S O C C H S O C C H S O C C H S O C C H S O C C H S O C C C H S O C C C C C C C C C C C C C C C C C C
,	Example No.	516	517	518	519	520	521

ما	CO2CH3	CO2CH3	CN	CN	N S	CM
(R)n	H	Ξ	×	Ξ.	æ	Ξ
TABLE 12 (continued)	S S S S S S S S S S S S S S S S S S S	CH ₃ L CH ₃ L CH ₃ CH ₄	(HOC2 H,)2 N- (HOCH-CH-CH-CH-CH-	(CH ₃ COC ₂ H ₄) ₂ N-•(-CH=CH-CH-CH-CH ₃ COC ₂ H ₄)	CH3 	- i i - сн з с с н, осси з
Example No.	522	523	524	525	526	527



	R	P. L. Q	CN, CN	CN, SO2CH3	CN, SO2C6H5	CH, CONIC, H,	CH, COMIC2H,OH	CN, CN	CN, SO ₂ CH ₃	CN, CO2CH3	CN, CO2CH3	-0=2-20- -0C-C=C=	-3 = 3 - 30 - 0
TABLE 13	$Q = HC - \frac{1}{1} \qquad \qquad$	<u>R</u> 1	C2H4OH	C2H4OH	C2H4OH	C2H,OH	C2H,OH	сн, сн, оссн,	CH ₂ CH ₂ OCCH ₃	си, си, оёси,	CH, CH, OU-C, H,	си, си, осси,	CH2CH(OH)CH3
		<u>(R)</u> n	=	=	=	=	æ	æ	Ħ	4,8-d1-CH3	8 -CH ₃	8 - CH 3	8 - CH 3
		Example No.	532	533	534	535	536	537	538	539	540	541	C P Y

P. Q. CONH, Cells	- 0.0- 0	CN, CO2CH3	CN, CN	CN, CO2CH3	CN, CO ₂ CH ₃	CN, CN	•=•	·	E 10.00 M		CN, CO ₂ CH ₃ CN, CO ₂ CH ₃	CN, CO ₂ CH ₃	CN, CO, CH3	1
TABLE 13 (continued) $\frac{R1}{C}$ Q	CH2CH2OCH2CH2OCH3		CH2	CoHs	CH2	CH ₂	0 0 0 0 0 0 0 0			CH ₂ CH ₂ N(CH ₃)SO ₂ CH ₃	CH2CH2OUNICEHS	CH2CH2CI	CH ₂ CH ₂ OC ₆ H ₅	CH2CH2SC6H5
(R)n	2-C1-8-CH ₃	4-C1-8-CH ₁	8-CH3	8-CH,	n	4-OCH,-8-CH,	HOTO TELL	1,4-10-1-1-0-1-1-0-1-1-0-1-1-1-1-1-1-1-1-1	==	±	6,8-di-CH3	6,9-di-CH ₃	8-CH3	B-CH3
Example No.	543	544	545	546	547	548		549	550	551	552	553	554	555

P. Q	CN, COOH	CN, CO2CH3	CN, CO2CH3	CN, CO2CH3	CN, CO ₂ CH ₃		(N) COSCH3	CN COCICHIAN	110 CO NO	(N) (O) (H)		CN, CO2CH3	CN, CN,
TABLE 13 (continued) \overline{R}_1	CH2CH2S-CS	CH2CH2SO2	CH ₂ CH ₂ N ii	-CH2CH2N ii	-CH2CH=CH2	-CH2CH2OC2H4OH	-CH2CH2SO2CH=CH2	-CH ₂ CH(OH)CH ₂ OH	-CH2C6H11	-CH2CH2SO2N(CH1)2	-CH2CH2CON(CH3)C&H5	-сн2сн2осн2с1	си,си,осси,
(R)n	4-0CII3-8-CH3	8 - CH 3	8-CH3	B-CH ₃	8-CH3	8-CII.	8-CH3	8 - CH 3	9 - CH3	9 CH3	9-CH3	4-CH3	4-CH3
Example No.	556	557	558	559	260	561	562	563	564	565	266	567	568

		P. 1.9	CN, CM	CN, SO2CH3	CN, SO2C6Hs	CN, CONHC&HS	CN, CONHC, II, OH	CN, CN	CN, SO ₂ CH ₃	CN, CO2CH3	CN, CO2CH3
TABLE 14	$C = HC - \begin{cases} 10 & 1 & 2 \\ 0 & 0 & 0 \end{cases}$ $R_1 = \begin{cases} R \\ R \\ R \end{cases}$	$\frac{R}{1}$	C ₂ H ₄ OH	C2H4OH	HOOD	CH1	C ₂ H ₄ OH	CH ₂ CH ₂ OCCH ₃	CH2CH2OCCH3	CH2CH2OCCH3	CH2CH2OC-C6H5
		(R)n	æ	I	≖	Ħ	×	æ	Ŧ	8-CH3	8-CH3
		Example No.	574	575	576	577	578	579	580	581	582

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	P. Q. O. C. H.	-0=0-00-	-06-c==c-	0 соин, с.н.s	CN, CO2CH3	CN, CN	CM, CO2CH3	CN, CO2CH3	CH, CN	-=- 	CN, CO2CH3	CM, CO,CH3	CN, CO2CH3
TABLE 14 (continued)	R1	CH ₂ CH ₂ OCCH ₃	CH2CH(OH)CH1	CH 2CH 2OCH 2CH 2OCCH 3	HOOD-	CH2	CeHs	CH ₂ COOH	CH2	си 2 си 2 оёси 3	HOOD-	CH2CH2N(CH3)SO2CH3	CH, CH, OCHIC, H5
ΔT	(R)n	8 - CH 3	8 - CH	2-C1-8-CH3	4-C1-8-CH3	5,8-di-CH3	Œ	=	=	3-CH3	3-CH3	3-CH3	3-0CH ₃ -8-CH ₃
	xample No.	583	8 ብ	585	586	587	588	589	590	591	592	593	594

Р, Р Си, СО ₂ СИ ₃ Си, СО ₂ СИ ₃	CN, COOH	CN, CO2CH3	CN, CO2CH3	CN, CO ₂ CH ₃
TABLE 14 (continued) 8.1 CH 2 CH 2 C 1 CH 2 CH 2 C 1 CH 2 CH 2 C 2 6 H 5	CH2CH2S-CS-CH3	CH ₂ CH ₂ N ii	-CH2CH2N ii i	-CH ₂ CH=CH ₂ -CH ₂ CH ₂ OC ₂ H ₄ OH -CH ₂ CH ₂ SO ₂ CH=CH ₂ -CH ₂ CH(OH)CH ₂ OH -CH ₂ CH(OH)CH ₂ OH -CH ₂ CH ₂ SO ₂ N(CH ₃) ₂ -CH ₂ CH ₂ CON(CH ₃) ₂
3-0CH ₃ -7-CH ₃ 3-0CH ₃ -7-CH ₃ 3-0CH ₃ -7-CH ₃	3,5-di-CH ₁ 3,5-di-CH ₁	3-0CH3	7,10-di-CH3	3,8-di-CH; 3,8-di-OCH; 8-OCH; 8-C1 8-CH; 8-CH;
Example No. 595 596	598	009	601	602 603 604 605 606 607

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P. 0	CN, CO ₂ CH ₃	CN, -CN,	CN, CN,	CN, -18	CN, CO2CH3	CN, CO2CH3	CN, CO2CH1
TABLE 14 (continued) R1	-CH,CH,OCCH,C1	-сн2сн2оссн3	-cH2OCH2	о -сн 2с н2оссн 3	CH2CH2N C-CH3	CH2CH2N C-S	CH2CH2-S-
(R)n	Ξ	œ	æ	5,8-di-CH3	5,8-di-CH3	8-CH3	7,9-d1-0CH3
Example No.	609	610	611	612	613	614	615

CN, CO2CH1

P, Q

о соон Свия -00-0-1

CO2CH1, CO2CH1

$$C = HC - \frac{1}{11} \frac{N}{9}$$

$$C = HC - \frac{1}{10} M$$

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C. Hs

CH3

618

CN, CD2CH2CH2OH

CN, CO2CH3

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0 1d	C C C B S C C C C C C C C C C C C C C C	0 CO2CH3 -OC-C===C	CN, CONHC2H,OH	O CM C& H s	CN, CO2CH3	CN, SO2	0 CO2CH3 C6H5	ос-с——с -ос-с——с
m	HD S	H CH CH S		—————————————————————————————————————	-N -SC ₂ H ₅	-N-C-CH3	CH3 CH3	<i>Z</i> `\
(R)10	C. H p-CO2CH3	Сенр-соон	C. Hp-Br	C & H h - P - OCH a	CeH,-p-CH1	C. H o - C. 1	C&H,-m-CO2CH3	C.Hp-C01CH1
Example No.	621	622	623	624	625	626	627	628

TABLE 15 (continued)

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0'4	CN, CO2CH,	CM, CONHC, H, OH		CM, CO2CH3	CN, CONH	0 CN C.H.s	-0C-C=C	CN, -CC.1
TABLE 15 (continued) B	—N Ц—S-С, Н, ОН	-N 0 0 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1			, = · \ , = ·	-CCH 3 C-CO 2C 2H 3	-N- 	-N- -II- S
(R)10	C. e. H. s	C 6 H 5	C ₆ H ₅	Cells	C ₆ H ₅	CeHs	CeHs	CeHs
Example No.	629	630	631	632	633	634	635	636

The preparation of the unsaturated polvesters of this invention is according to well known techniques in the art as described, for example, in U.S. Patents: 3.642,672; 3,549,586; 4,299,927; and 4,355,136. It is preferred that these polyesters have an acid number of from about 10 to about 28, an inherent viscosity of from about 0.05 to about 0.25, and a number average molecular weight of from about 1100 to about 3800. Typical colored polyester material prepared in accordance with this invention are as follows:

Example 637

Preparation of Unsaturated Polyester Material from Neopentyl Glycol, isophthalic Acid, and Maleic Anhydride Copolymerized with Methine Colorant and Cured With Styrene

The reaction apparatus comprising a one-litre flask is fitted with a stirrer, thermometer, nitrogen inlet tube, and heated Vigreux column. The top of the Vigreux column is also fitted with a Dean-Stark trap and cold water condenser. The flask is charged with 251.6 g of neopentyl glycol (2.415 mol), 191.0 g of isophthalic acid (1.15 m), 0.0951 g (200 ppm) of the reactive methine compound

and 0.55 g of dibutyltin oxide. The flask is then heated to reflux and held at this temperature until the theoretical amount of distillate is collected from this esterification stage. The reaction system is cooled to 145°C and 112.8 g of maleic anhydride (1.15 m) and 0.055 g of toluhydroquinone are added. The esterification and polycondensation reactions are continued for one hour at 175°C, then for one hour at 185°C, and then at 195°C until an acid number of 18.1 for the unsaturated polyester is obtained. The inherent viscosity of this polyester is 0.133, the number average molecular weight is 2640 and the color is bright red-orange. Sufficient of this polyester material is blended or dissolved in styrene monomer to give 40 wt.% monomer and the system then blended with 1 wt% benzoyl peroxide. One-eighth inch thick sheet castings are prepared by decanting the blend between glass plates separated by i-inch thick gasket. The blend is cured by heating for two hours at 70°C, then for one hour at 100°C, then for two hours at 125°C, and then for one hour at 150°C. After cooling, the cured polyester sheet is removed and cut into bars. The flexural strength of the bars is 18.46×10^3 psi (127 N/mm²).

Example 638

Preparation of Unsaturated Polyester Material from Propylene Glycol, Isophthalic Acid, and Maleic Anhydride Copolymerized With Methine Colorant and Cured With Styrene

The following components are charged into the equipment described in Example 801:

192.5 g propylene glycol (2.53 mol); 191.0 g isophthalic acid (1.15 mol); 0.50 g dibutyltin oxide;) and

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These components are reacted as described in Example 801 until the theoretical amount of distillate is collected. The reaction is cooled to 145°C and 112.8 g maleic anhydride (1.15 mol) and 0.05 g of toluhydroquinone are added. The reaction is continued for one hour at 175°C, then for one hour at 185°C, and then at 195°C until an acid number of 14.1 for the unsaturated polyester is obtained. The inherent viscosity of this polyester is 0.11, the number average molecular weight is 1912, and the color is yellow. A curable blend of this unsaturated polyester and sheet casting and bars thereof were prepared as in Example 801. The bars had a flexural strength of 18.60×10^3 psi (128 N/mm²).

The above inherent viscosities, acid numbers, number average molecular weights, and fiexural strengths were determined as follows:

Acid number by ASTM D-1639-70;

Number Average Moplecular Weight by ASTM D-08.03 (Gel Permeation Chromatography); Fiexural Strength by ASTM D-790-81;

Inherent Viscosity according to ASTM D2857—70 procedure in a Wagner Viscometer of Lab Glass Inc. of Vineland, N.J. having a 1/2 ml capitary bulb, using a polymer concentration of 0.5% by weight in 60/40 by weight, phenol/tetrachloroetnane solvent. The procedure comprises heating the polymer/solvent system at

120°C for 15 minutes to enhance dissolution of the polymer, cooling the solution to 25°C and measuring the time of flow at 25°C. The LV, is calculated from the equation

$$\begin{array}{ccc}
25^{\circ}C & & & \frac{t_s}{-} \\
(\eta) & = & \frac{t_o}{-} \\
0.50\% & & & C
\end{array}$$

where:

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 $\langle \eta \rangle$ = Inherent viscosity at 25°C at a polymer concentration of 0.5 g/(00 ml of solvent;

In = Natural logarithm;

t_s = Sample flow time;

to = Solvent-blank flow time; and

C = Concentration of polymer in grams per 100 ml of solvent = 0.50.

Claims

1. A colored composition comprising unsaturated polyester material, wherein the polyester has an I.V. of 0.05 to 0.25, an acid number of 10 to 28, and a number average molecular weight of from 1100 to 3800, 20 having copolymerized therein a total of from 1.0 to 5000 ppm of at least one methine moiety, said moiety absorbing in the range of from 320 nm to 650 nm and being nonextractable from said polyester material, wherein each methine moiety is derived from a reactive compound having the formula

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$$P$$
 $C=CH-A$
,
 $A-CH=C P$
 $C=CH-A$
,
 (R)

wherein each A is selected from the following radicals:

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55 $(R) = \begin{pmatrix} R \\ R \end{pmatrix}_{n} \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \end{pmatrix}_{n} \begin{pmatrix} R \\ R \end{pmatrix}_{n} \end{pmatrix}_{n$ 60

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$$R_4$$
 R_5
 R_6
 R_1
 R_{10}
 R_{10}

wherein:

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R and R' are selected from hydrogen, fluorine, chlorine, bromine, alkyl, alkoxy, phenyl, phenoxy, alkyltic, and arylthio; r. is 0, 1, 2;

 R_1 and R_2 are selected from hydrogen; cycloalky); cycloalky, substituted with one or two of alky, —OH, alkoxy, halogen, or nydroxy substituted alkyl; phenyl; phenyl; phenyl substituted with alkyl, alkoxy, halogen, alkanoylamino, carboxy, cvano, or alkoxycarbony; straight or branched lower alkenyl; straight or branched alkyl of 1—8 carbons and such alkyl substituted with the following: hydroxy; halogen; eyano; succinimido; hydroxysuccinimido; acyloxysucinimido; glutarimido; phenylcarbamoyloxy; pnthalimido; 4carboxyphthalimido; pnthalimidino; 2-pyrrolidono; cyclohexyl; phenyl; pnenyl substituted with alkyl, alkoxy, halogen, hydroxy alkanoylamine, carboxy, cyano, or alkoxycarbonyl; alkylsulfamoyl; vinylsulfonyl; pnenylsulfonamido; alkylsulfonamido; benzoylsulfonicimido; sulfamyl; alkoxycarbonylamino; alkylcarbamoyloxy; alkoxycarbonyl; alkoxycarbonyloxy; alkenylcarbonylamino; groups of the formula

wherein Y is --NH--,

-O-, -S-, or $-CH_2O-$; $-S-R_{14}$; $SO_2CH_2CH_2SR_{14}$; wherein R_{14} is alkyl, phenyl phenyl substituted with halogen, alkyl, alkoxy, alkanoylamino, cyano, or alkoxycarbonyl; pyridyl; pyrimidinyl; benzoxazolyl; 25 benzimidazolyl; benzothiazolyl; radicals of the formulae

 $-OSR_{16}$; $-NHXR_{16}$; $-X-R_{16}$; $-CONR_{15}R_{15}$; and $-SO_2NR_{15}R_{16}$; wherein R_{15} is selected from H, aryl, alkyl, and alkyl substituted with halogen, -OH, phenoxy, aryl, -CN, cycloalkyl, alkylsulfonyl, alkylthio, alkanoyloxy, or alkoxy; X is —CO—, —COO—, or —SO₂—; R₁₆ is selected from alkyl and alkyl substituted with halogen, hydroxy, phenoxy, aryl, cyano, cycloalkyl, alkylsulfonyl, alkylthio, alkanoyloxy, and alkoxy; and when X is —CO—, R₁₆ also can be hydrogen, amino, alkenyl, alkylamino, dialkylamino, arylamino, aryl, or furyl; alkoxy; alkoxy substituted with hydroxy, cyano, alkanoyloxy, or alkoxy; phenoxy; phenoxy substituted with one or more of alkyl, carboxy, alkoxy, carbalkoxy, or halogen; R_1 and R_2 can be a single combined group such as pentamethylene, tetramethylene, ethyleneoxy ethylene, ethylene sulfonylethylene, or

which, with the nitrogen to which it is attached, forms a ring; R₁₇ is alkyl, aryl, or cycloalkyl;

 R_3 is alkylene, arylene, aralkylene, alkyleneoxy, or alkyleneoxyalkylene;

Z is a direct single bond, OCO, O, S, SO₂, R₁₇SO₂N=,

arylene, or alkylene;

 R_4 , R_5 , and R_6 are each selected from hydrogen and alkyl;

R₇ is carboxy, carbalkoxy, or (R),

Ric is hydrogen, alkyl, and aryl;

 R_{ϵ} and R_{ϵ} are each selected from hydrogen and substituted or unsubstituted alkyl, aryl, or cycloalkyl;

 R_{11} and R_{12} are each selected from hydrogen, alkyl, hydroxyl, or acyloxy;

B represents the atoms necessary to complete a five or six membered ring and is selected from

each P and Q are selected from cyano, carbalkoxy, carbaryloxy, carbaralkyloxy, carbamyl, carboxy, Nalkylcarbamyı, N-alkyl-N-arylcarbamyl, N,N-dialkylcarbamyl, N-arylcarbamyl, N-cyclohexylcarbamyl, aryl, $\hbox{2-benzoxazolyl, 2-benzothiazolyl, 2-benzimidazolyl, 1,3,4-thiadiazol-2-yl, 1,3,4-oxadiazol-2-yl, SO_2 alkyl, S$ aryl, and acyl, or P and Q may be combined as

wherein R_{17} is defined above and R_{18} is CN, COOH, CO_2 alkyl, carbamyl, or N-alkylcarbamyl;

wherein at least one of A, P, and Q for each dye molecule must be or bear a condensable group selected from carboxy, carbalkoxy, carbaryloxy, N-alkylcarbamyloxy, acyloxy, chlorocarbonyl, alkylamino, hydroxyl, N-phenylcarbamyloxy, N-(alkyl)₂carbamyloxy, amino, carbamyloxy, cyclohexanoyloxy, and carbocyclohexyloxy; and

wherein in the above definitions, each alkyl, aryl, or cycloalkyl moiety or portion of a group or radical may be substituted where appropriate with hydroxyl, acyloxy, alkyl, cyano, alkoxycarbonyl, halogen, alkoxy, or aryl, aryloxy, or cycloalkyl.

- 2. The composition of claim 1 wherein the polyester acid component is comprised of 40—60 mole% isophthalic acid and conversely 60—40 mole% of either or a mixture of maleic fumaric acid, and the alcohol component is comprised of propylene glycol, neopentyl glycol, or mixtures thereof.
- 3. The composition of claim 3 wherein the polyester acid component is comprised of isophthalic acid and maleic acid, and the alcohol is neopentyl glycol or neopentyl glycol mixed with less than about 75 moie% propylene glycol.
 - 4. The composition of claim 1 wherein the reactive compound has the formula

5. The composition of claim 4 wherein:

R is H, alkyl, halogen or alkoxy; and

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 $R_{
m s}$ and $R_{
m g}$ are each selected from H, alkyl, cycloalkyl, cycloalkyl substituted with one or more of alkyl, OH, CN, alkoxy, carbalkoxy or alkanovioxy, and alky: substituted with one or more of OH, CN, alkanovioxy, carbalkoxy, aryl, substituted aryl, alkoxy, alkoxyalkoxy, nalogeri, succinimiod or carbamyl.

6. The composition of claim 1 wherein the reactive compound has the formula

7. The composition of claim 6 where:

R is H, alkyl, halogen or alkoxy;

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 R_1 is selected from H, alkyl, cycloalkyl, cycloalkyl substituted with one or more of alkyl, OH, CN, alkoxy, carbalkoxy or alkanoyloxy, and alkyl substituted with one or more of OH, CN, alkanoyloxy, carbalkoxy, aryl, substituted aryl, alkoxy, alkoxy-alkoxy, halogen, succinimido or carbamyl; and

 R_4 , R_5 and R_6 are each H or alkyl.

8. The composition of claim 1 wherein the reactive compound has the formula

9. The composition of claim 8 wherein:

R and R' are each selected from H, alkyl, halogen and alkoxy; and

R₁ is H, alkyl, cycloalkyl, cycloalkyl substituted with one or more of alkyl, OH, CN, alkoxy, carbalkoxy or alkanoyloxy, and alkyl substituted with one or more of OH, CN, alkanoyloxy, carbalkoxy, aryl, substituted aryl, alkoxy, alkoxyalkoxy, halogen, succinimido or carbamyl.

10. The composition of claim 1 wherein the reactive compound has the formula

11. The composition of claim 10 wherein:

each R is H, alkyl, halogen or alkoxy; each R₁ is H, alkyl, cycloalkyl, cycloalkyl substituted with one or more of alkyl, OH, CN, alkoxy, carbalkoxy or alkanoyloxy, and alkyl substituted with one or more of OH, CN, alkanoyloxy, carbalkoxy, aryl, substituted aryl, alkoxy, alkoxyalkoxy, halogen, succinimido or carbamyl;

12. The composition of claim 1 wherein the condensable group is one or more of carboxy, carbalkoxy

13. The composition of claim 1 wherein the reactive compound is

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14. The composition of claim 1 wherein the reactive compound is

$$(\mathsf{CH}_3)_2 \mathsf{N} - (\mathsf{CH}_3)_2 \mathsf{N} - (\mathsf{CH}_3)_2$$

15. The composition of claim 1 wherein the reactive compound is

16. The composition of claim 1 wherein the reactive compound is

17. The composition of claim 1 wherein the reactive compound is

$$(CH_3COCH_2CH_2)_2-N-CH=CH_3$$

18. The composition of claim 1 wherein the reactive compound is

- 19. The composition of any of claims 1-3 blended with a curing agent.
- 20. The cured composition of claim 19.
- 21. Formed articles of the cured composition of claim 20.

Patentansprüche

1. Farbige Zusammensetzung mit einem ungesättigten Polyestermaterial, in der der Polyester eine I.V. von 0,05 bis 0,25, eine Säurezahl von 10 bis 28 sowie eine mittlere Molekulargewichtszahl von 1100 bis 3800 hat, und der eincopolymerisiert insgesamt 1,0 bis 5000 ppm mindestens einer Methingruppierung enthält, die im Bereich von 320 nm bis 650 nm absorbiert und aus dem Polyestermaterial nicht extrahierbar ist, wobei jede Methingruppierung sich von einer reaktiven Verbindung mit der Formel

P
$$P = CH-A$$
 , $A-CH=C P = CH-A$, $P =$

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15 ableitet, wobei jedes A aus einem der folgenden Reste ausgewählt ist:

$$(R) \xrightarrow{n} \underset{R_{1}}{\overset{0}{\downarrow}} (R)_{n} ,$$

$$(R) = \begin{pmatrix} R \\ R \end{pmatrix} = \begin{pmatrix} R \\ R \end{pmatrix}$$

$$\begin{array}{c|c} & & & & \\ & & & \\ \hline \vdots & & & \\ \hline R_7 & & CH_3 \\ \hline CH_3 & & & \\ \hline \end{array} \qquad \begin{array}{c} & & \\ R_2 \\ \hline R_1 & & \\ \hline \end{array} \qquad \begin{array}{c} & - CH = CH - \\ \hline \end{array} \qquad \begin{array}{c} & \\ & \vdots \\ \hline \end{array} \qquad \begin{array}{c} & - CH = CH - \\ \hline \end{array} \qquad \begin{array}{c} & \\ & \\ \end{array} \qquad \begin{array}{c} & \\ \\ \end{array} \qquad$$

$$\begin{array}{c|c} R_4 & R_5 \\ \vdots & \vdots & \ddots & R_6 \\ (R)_n & R_1 & R_{10} \end{array}$$

worin:

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:0

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R und R' ausgewählt sind aus Wasserstoff, Fluor, Chlor, Brom, Alkyl, Alkoxy, Phenyl, Phenoxy, Alkylthio und Arylthio; und n gleich 0, 1 oder 2 ist;

R₁ und R₂ ausgewählt sind aus Wasserstoff, Cycloalkyl; Cycloalkyl, das substituiert ist durch ein oder zwei Alkyl, OH-, Alkoxy, Halogen, oder Hydro-substituiertem Alkyl; Phenyl; Phenyl, substituiert durch Alkyl, Alkoxy, Halogen, Alkanoylamino, Carboxy, Cyano oder Alkoxycarbonyl; geradkettigem oder verzweigtkettigem kurzkettigen Alkenyl; geradkettigem oder verzweigtkettigem Alkyl mit 1-8 Kohlenstoffatomen und solchem Alkyl, das substituiert ist durch: Hydroxy; Halogen; Cyano; Succinimido; Hydroxysuccinimido; Acyloxysuccinimido; Glutarimido; Phenylcarbamoyloxy; Phthalimido; 4-Carboxyphthalimido; Phthalimidino; 2-Pyrrolidono; Cyclohexyl; Phenyl; Phenyl substituiert mit Alkyl, Alkoxy, Halogen, Hydroxyalkanoylamino, Carboxy, Cyano oder Alkoxycarbonyl; Alkylsulfamoyl; Vinyisulfonyl; Acrylamido; Sulfamyl, Benzoyisulfonicimido; Alkylsulfonamido, Phenylsulfonamido; Alkoxycarbonylamino, Alkylcarbamoyloxy; Alkoxycarbonyl; Alkoxycarbonyloxy; Alkenylcarbonylamino; Gruppen der Formel

in der Y gleich —NH—, —N-alkyl, —O—, —S—, oder —CH $_2$ O— ist; —SR $_{14}$; SO $_2$ CH $_2$ CH $_2$ SR $_{14}$; worin R $_{14}$ gleich Alkyl, Phenyl, Phenyl substituiert mit Halogen, Alkyl, Alkoxy, Alkanoylamino, Cyano oder Alkoxycarbonyl ist; Pyridyl; Pyrimidinyl; Benzoxazolyl, Benzimidazolyl; Benzothiazolyl; Resten der Formeln

 $-OXR_{16}$; $-NHXR_{16}$; $-X-R_{16}$; $-CONR_{15}R_{15}$; und $-SO_2NR_{15}R_{15}$; worin R_{15} ausgewählt ist aus H, Aryl, Alkyl, und Alkyl substituiert mit Halogen, —OH, Phenoxy, Aryl-, —CN, Cycloalkyl, Alkylsulfonyl, Alkylthio, Alkanoyloxy oder Alkoxy; X ist gleich —CO—, —COO—, oder —SO₂—, R₁₆ ist, ausgewählt aus Alkyl und Alkyl, substituiert mit Halogen, Hydroxy, Phenoxy, Aryl, Cyano, Cycloalkyl, Alkylsulfonyl, Alkylthio, Alkanoyloxy, und Alkoxy, wobei gilt, daß wenn X gleich —CO— ist, R₁₆ ferner stehen kann für Wasserstoff, Amino, Alkenyi, Alkylamino, Dialkylamino, Arylamino, Aryl oder Furyl; Alkoxy; Alkoxy, substituiert mit Hydroxy, Cyano, Alkanoyloxy oder Alkoxy; Phenoxy; Phenoxy, substituien mit einem oder mehreren Alkyl, Carboxy, Alkoxy, Carbalkoxy oder Halogen; und worin R, und R2 eine einzelne kombinierte Gruppe bilden konnen, z.B. Pentamethylen, Tetramethylen, Ethylenoxyethylen, Ethylensulfonylethylen oder

XR₁₇

Ethylen-N-ethylen,

die mit dem Stickstoff, an den sie gebunden ist, einen Ring bildet, wobei R₁₇ für Alkyl, Aryl oder Cycloalkyl steht;

 R_s steht für Alkylen, Arylen, Aralkylen, Alkylenoxy oder Alkylenoxyalkylen; Z ist eine direkte einfasne Bindung oder OCO, O, S, SO₂, R_1 -SO₂N=,

Arylen oder Alkylen;

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 $R_4,\,R_5$ und R_6 sind jeweils ausgewählt aus Wasserstoff und Alkyl;

R7 steht für Carboxy, Carbalkoxy oder (R),

R₁₀ ist Wasserstoff, Alkyl oder Aryl;

R_s ud R_s sind ausgewählt aus Wasserstoff und substituiertem oder unsubstituiertem Alkyl, Aryl oder Cycloalkyl;

R₁₁ und R₁₂ sind jeweils ausgewählt aus Wasserstoff, Alkyl, Hydroxyl oder Acyloxy;

B steht für die Atome, die erforderlich sind, um einen fünf- oder sechsgliedrigen Ring zu vervollständigen und ist ausgewählt aus:

P und Q sind jeweils ausgewählt aus Cyano, Carbalkoxy, Carbaryloxy, Carbaralkyloxy, Carbamyl, Carboxy, N-Alkylcarbamyl, N-Alkyl-N-arylcarbamyl, N-N-Dialkylcarbamyl, N-Arylcarbamyl, N-Cyclohexylcarbamyl, Aryl. 2-Benzoxazolyl, 2-Benzothiazoyl, 2-Benzimidazolyl, 1,3,4-Thiadiazol-2-yl, 1,3,4-Oxadiazol-2-yl, SO₂-Alkyl, SO₂-Aryl und Acyl oder P und Q können kombiniert sein zu

worin R_{17} die bereits angegebene Bedeutung hat und R_{18} steht für-CN, —COOH, —CO₂-alkyl, Carbamyl oder N-alkylcarbamyl;

wobei mindestens einer der Reste A, P und Q für jedes Farbstoffmolekül eine kondensierbare Gruppe,

ausgewählt aus den folgenden Gruppen sein muß oder eine solche Gruppe tragen muß: Carboxy, Carbalkoxy, Carbaryloxy, N-Alkyloarbamyloxy, Acyloxy, Chiorocarbonyl, Carbamyloxy, N-(Alkylizcarpamyloxy, Amino, Alkylamino, Hydroxyl, N-Phenylcarbamyloxy, Cyclonexanoyloxy und Carbocyclonexyloxy;

und worin in den angegebenen Definitionen jeder Alkyl-, Aryl- oder Cycloalkylrest gegebenenfalls substituien sein kann mit Hydroxyl, Acyloxy, Alkyl, Cyano, Alkoxycarbonyl, Halogen, Alkoxy oder Aryl, Aryloxy oder Cycloalkyl.

- 2. Zusammensetzung nach Anspruch 1, in der die Polyester-Säurekomponente zu 40-60 Mol-% aus lsophthalsäure und zu 60-40 Mol-% aus entweder Maleinsäure oder Furnarsäure oder einer Mischung hiervon besteht und die Alkoholkomponente aus Propylengiykol, Neopentyligiykol oder einer Mischung hiervon.
- 3. Zusammensetzung nach Anspruch 2, in der die Polyester-Säurekomponente aus Isophthaisäure und Maleinsaure und die Alkoholkomponente aus Neopentyiglykol, oder Neopentyigiykol, vermischt mit weniger als etwa 75 Mol-% Proylengiykol besteht.
 - 4. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung die folgende Formel hat;

5. Zusammensetzung nach Anspruch 4, in der bedeuten:

R gleich Wasserstoff, Alkyl, Halogen oder Alkoxy und

R₁ und R₂ jeweils ausgewählt sind aus Wasserstoff, Alkyl, Cycloalkyl, Cycloalkyl substituiert mit einem oder mehreren Alkyl, —OH, —CN, Alkoxy, Carbalkoxy, oder Alkanoyloxy und Alkyl substituiert mit einem oder mehreren —OH, —CN, Alkanoyloxy, Carbalkoxy, Aryl, substituiertem Aryl, Alkoxy, Alkoxyalkoxy, Halogen, Succinimido oder Carbamyl.

6. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung der folgenden Formel entspricht:

7. Zusammensetzung nach Anspruch 6, worin bedeuten:

R Wasserstoff, Alkyl, Halogen oder Alkoxy;

R₂ Wasserstoff, Alkyl, Cycloalkyl, Cycloalyl substituiert mit einem oder mehreren Alkyl, —OH, —CN, Alkoxy, Carbalkoxy oder Alkanoyloxy und Alkyl substituiert mit einem oder mehreren -OH, -CH, Alkanoyloxy, Carbalkoxy, Aryl, substituiertem Aryl, Alkoxy, Alkoxy-alkoxy, Halogen, Succinimido oder Carbamyl und

 R_4 , R_5 und R_6 jeweils Wasserstoff oder Alkyl.

8. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung der folgenden Formel entspricht:

9. Zusammensetzung nach Anspruch 8, in der bedeuten:

R und R' jeweils Wasserstoff, Alkyl, Halogen oder Alkoxy und

R₁ Wasserstoff, Alkyl, Cycloalkyl, Cycloalkyl substituiert mit einem oder mehreren Alkyl-, —OH, —CN, Alkoxy, Carbalkoxy oder Alkanoyloxy und Alkyl substituiert mit einem oder mehreren --OH, --CN, Alkanoyloxy, Carbalkoxy, Aryl, substituiertem Aryl, Alkoxy, Alkoxyalkoxy, Halogen, Succinimido oder Carbamyl.

10. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung der folgenden Formel entspricht:

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11. Zusammensetzung nach Anspruch 10, worin bedeuten

R jeweils Wasserstoff, Alkyl, Halogen oder Alkoxy;

R₁ jeweits Wasserstoff, Alkyl, Cycloalkyl, Cycloalkyl substitutert mit einem oder mehrerer, Alkyl, —OH, —CN, Alkoxy, Carbalkoxy oder Alkanovioxy und Alkyl substituier, mit einem oder mehreren —OH, —CN, Alkanovioxy, Carbalkoxy, Aryl, substituiertem Aryl, Alkoxy, Alkoxyalkoxy, Halogen, Succinimide oder Carbamyl; und

12. Zusammensetzung nach Anspruch 1, in der die kondensierbare Gruppe aus einer oder mehreren Carboxy-, Carbalkoxy- oder Hydroxygruppen besteht.

13. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung besteht aus:

14. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung besteht aus:

$$(CH_3)_2N^{-1}$$
 $CH_2C_2C_2H_5$

15. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung besteht aus:

$$\begin{array}{c}
\text{NC} \\
\text{CH}_{3}\text{O}_{2}\text{C}
\end{array}$$

$$\begin{array}{c}
\text{CH}_{3}\text{O}_{2}\text{C}
\end{array}$$

$$\begin{array}{c}
\text{CH}_{3}\text{O}_{2}\text{C}
\end{array}$$

$$\begin{array}{c}
\text{CH}_{3}\text{O}_{2}\text{C}
\end{array}$$

$$\begin{array}{c}
\text{CH}_{3}\text{O}_{2}\text{C}
\end{array}$$

16. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung besteht aus:

17. Zusammensetzung nach Anspruch 1, in der die reaktive Verbindung besteht aus:

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18. Zusammensetzung nach Ansprück 1, in der die reaktive Verbindung besteht aus:

- 19. Zusammensetzung nach einem der Ansprüche 1—3, vermischt mit einem Härungsmittel.
- 20. Die gehärtete Zusammensetzung nach Anspruch 19.
- 21. Formkörper, hergestellt aus der gehärteten Zusammensetzung nach Anspruch 20.

Revendications

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1. Composition colorée comprenant un polyester insaturé, ayant une viscosité inhérente comprise entre 0,05 et 0,25, un indice d'acidité compris entre 10 et 28 et un poids moléculaire moyen en nombre compris entre 1100 et 3800, dans lequel est copolymérisé un total de 1,0 à 5000 ppm d'au mons un groupement méthine, ledit groupement méthine absorbant dans la région du spectre comprise entre 320 nm et 650 nm et étant non extractible dudit polymère, et dans lequel chaque groupement méthine est dérivé d'un composé réactif ayant la formule:

PC=CH-A

QC=CH-A

$$A-CH=C R_1$$
 R_1
 R_1

où chaque A est choisi parmi les radicaux suivants:

(R)
$$\stackrel{|}{\underset{R}{\longrightarrow}}$$
 (R) $\stackrel{|}{\underset{R}{\longrightarrow}}$ (R)

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où:

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R et R' sont choisis parmi les atomes d'hydrogène, de fluor, de chlore, de brome, les radicaux alkyle, alkoxy, phényle, phénoxy, alkylthio, et arylthio; n est 0, 1, 2;

 R_1 et R_2 sont choisis parmi un atome d'hydrogène; un radical cycloalkyle; cycloalkyle substitué par un ou deux alkyle, —OH, alkoxy, halogène ou alkyle substitué par un groupe hydroxy; un radical phényle; phenyle substitué par un alkyle, alkoxy, halogène, alkanoylamine, carboxy, cyano, ou alkoxycarbonyle; un radical alkènyle inférieur droit ou ramifié; un radical alkyle droit ou ramifié de 1 à atomes de carbone et un radical alkyle substitue par les groupes suivants; hydroxy; halogène; cyanc; succinimido; hydroxysuccinimido; acyloxysuccinimido; glutarimido; phénylcarbamoyloxy; phtalimido; 4carboxyphtalimido; phtalimidono; 2-pyrrolidono; cyclohexyle; phényle; phényle substitué par un groupe alkyle, alkoxy, halogène hydroxy alkanoylamino, carboxy, cyano, ou alkoxycarbonyle; alkylsulfamoyle;

vinyisulfonyie; acrylamido; sulfamyie; benzovisulfonicimido; alkylsulfonamido; phenyisulfonamido; alkoxycarbonylamino; alkyicarbamoyloxy; alkoxycarbonyie; alkoxycarbonyioxy; alkenyicarbonylamino; les groupes de formule:

où Y est —NH—, —N-alkyle, —C—, —S—, ou —CH2O—; —S—R14; $SO_2CH_2CH_2SR_{12}$; où R14 est un radical alkyle, phényle, phényle substitué par un halogène, alkyle, alkoxy, alkanoylamino, cyano, ou alkoxycarbonyle; pyridyle; pyrimidinyle; benzoxazolyle; benzimidazolyle; benzothiazolyle; ou un radical de formule:

 $--\mathsf{OXR}_{16}; \ --\mathsf{NHXR}_{16}; \ --\mathsf{X}--\mathsf{R}_{16}; \ --\mathsf{CONR}_{15}\mathsf{R}_{15}; \ \text{et} \ --\mathsf{SO}_2\mathsf{NR}_{15}\mathsf{R}_{15}; \ \text{où} \ \mathsf{R}_{15} \ \text{est choisi parmi un atome}$ d'hydrogène, un groupe aryle, alkyle, et alkyle substitué par un halogène, —OH, phénoxy, aryle, —CN, cycloalkyle, alkylsulfonyle, alkylthio, alkanoyloxy, ou alkoxy; X est -CO-, -COO-, ou $-SO_2-$; $R_{1\epsilon}$ est choisi parmi les radicaux alkyle et alkyle substitué par un halogène, hydroxy, phénoxy, aryle, cyano, cycloalkyle, alkylsulfonyle, alkylthio, alkanoyloxy, et alkoxy; et lorsque X est —CO—, R₁₆ peut aussi être un atome d'hydrogène, un radical amino, alkényle, alkylamino, dialkylamino, arylamino, aryle ou furyle; alkoxy; alkoxy substitué par un hydroxy, cyano, alkanoyloxy, ou alkoxy; phénoxy; phénoxy substitué par un ou plusieurs alkyle, carboxy, alkoxy, carbalkoxy, ou halogène; R1 et R2 peuvent former un groupe unique tel que pentaméthylène, tétraméthylène, éthylèneoxyéthylène, éthylène sulfonyléthylène, ou

qui, avec l'atome d'azote auquel il est attaché forme un cycle; R₁₇ est alkyle, aryle ou cycloalkyle;

R₂ est alkylène, arylène, aralkylène, alkylèneoxy, ou alkylèneoxyalkylène;

Z est une liaison simple, OCO, O, S, SO₂, R₁₇SO₂N=,

arylène, ou alkylène;

 $R_4,\,R_5,\,$ et R_6 sont chacun choisis parmi un atome d'hydrogène et un radical alkyle;

 R_7 est un radical carboxy, carbalkoxy, ou $(R)_n$;

R₁₀ est un atome d'hydrogène, un radical alkyle, et aryle;

 R_{B} et R_{S} chacun choisis parmi un atome d'hydrogène, un radical alkyle, aryle ou cycloaikyle substitué

 R_{11} et R_{12} sont chacun choisis parmi un atome d'hydrogène, un radical alkyle, hydroxyle ou acycloxy B représente le nombre d'atomes nécessaire pour compléter un cycle à 5 ou 6 chaînons et est choisi parmi;

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chacun des P et Q est choisi parmi les radicaux cyano, carbalkoxy, carbaryloxy, carbaralkyloxy, carbamyle, carboxy, N-alkylcarbamyle, N-alkyl-N-arylcarbamyle, N,N-dialkylcarbamyle, N-arylcarbamyle, Ncyclohexylcarbamyle, aryle, 2-benzoxazolyle, 2-benzothiazolyle 2-benzimidazolyle, 1,3,4-thiadiazol-2-yle, 1,3,4-oxadiazol-2-yle, SO₂-alkyle, SO₂-aryle, et acyle, ou P et Q peuvent être combinés sous la formule

où R_{17} est défini comme ci-dessus et R_{18} est CN, COOH, CO_2 -alkyle, carbamyle, ou N-alkylcarbamyle;

où au moins un des A, P, et Q pour chaque molécule de colorant peut être ou peut porter un groupe condensable choisi parmi les groupes carboxy, carbalkoxy, carbaryloxy, N-alkylcarbamyloxy, acyloxy, $\text{chiorocarbonyle,} \quad \text{carbamyloxy,} \quad \text{N-(alkyl)}_{2}\text{-carbamyloxy,} \quad \text{amino,} \quad \text{alkylamino,} \quad \text{hydroxyle,}$ phénylcarbamyloxy, cyclohexanoyloxy, et carbocyclohexyloxy; et

où dans les définitions ci-dessus, chaque groupement ou portion de groupement ou radical alkyle, aryle, ou cycloalkyle peut être substitué là où c'est possible par un radical hydroxyle, acyloxy, alkyle, cyano, alkoxycarbonyle, un atome d'halogène, un radical alkoxy, ou aryle, aryloxy, ou cycloalkyle.

2. Composition selon la revendication 1, dans lequel la composante acide du polyester comprend de 40 à 60% en poids d'acide isophtalique, et inversement 60 à 40% soit d'acide maléique soit d'acide fumarique, soit d'un mélange d'acide maléique et d'acide fumarique, et la composante alcool du polyester comprend du polypropylène glycol, du néopentyl glycol ou des mélanges de ces derniers.

3. Composition selon la revendication 2, dans lequel la composante acide du polyester comprend de l'acide isophtalique et de l'acide maléique et l'alcool est le néopentyl glycol, où du néopentyl glycol mélange à au moins d'environ 75% en moles de propylène glycol.

4. Composition selon la revendication 1, dans laquelle le composé réactif a la formule:

5. Composition selon la revendication 4, dans laquelle;

R est H, alkyle, halogène ou alkoxy; et

 R_1 et R_2 sont chacun séparément choisis parmi H alkyle, cycloalkyle, cycloalkyle substitué par un ou plusieurs groupes alkyle, OH, CN, alkoxy, carbalkoxy ou alkanoyloxy, et alkyle substitué par un ou plusieurs OH, CN, alkanoyloxy, carbalkoxy, aryle, aryle substitué, alkoxy, alkoxyalkoxy, halogène, succinimido ou carbamyle.

6. Composition selon la revendication 1, dans laquelle le composé réactif a la formule:

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7. Composition seion la revendication 6, dans laquelle;

R est H, alkyle, nalogène ou alkoxy;

R₃ est choisi parmi H, alkyle, cycloalkyle, cycloalkyle substitué par un ou plusieurs alkyle, OH, CN, alkoxy, carbalkoxy ou alkanoyloxy, et alkyle substitué par un ou plusieurs OH, CN, alkanoyloxy, carbalkoxy, aryle, aryle substitué, alkoxy, alkoxyalkoxy, nalogène, succinimido ou carbamyle; et

 $R_{4\ell}\,R_{\epsilon}$ et R_{ϵ} sont chacun H ou alkyle.

8. Composition selon la revendication 1, dans laquelle le groupe réactif a la formule:

9. Composition selon la revendication 10, dans laquelle R et R' sont chacun choisis parmi H, alkyle,

 R_1 est H, alkyle, cycloalkyle, cycloalkyle substitué par un ou plusieurs alkyle, OH, CN, alkoxy, carbalkoxy halogène et alkoxy; et ou alkanoyloxy, et alkyle substitué par un ou plusieurs OH, CN, alkanoyloxy, carbalkoxy, aryle, aryle substitue, alkoxy, alkoxyalkoxy, halogène, succinimido ou carbamyle.

10. Composition selon la revendication 1, dans laquelle le composé réactif a la formule:

11. Composition selon la revendication 10, dans laquelle chaque R est H, alkyle, halogène ou alkoxy; 30 chaque R₁ est H, alkyle, cycloalkyle, cycloalkyle substitué par un ou plusieurs alkyle, OH, CN, alkoxy, carbalkoxy ou alkanoyloxy, et alkyle substitué par un ou plusieurs OH, CN, alkanoyloxy, carbalkoxy, aryle, aryle substitué, alkoxy, alkoxyalkoxy, halogène, succinimido ou carbamyle, et

12. Composition selon la revendication 1, dans laquelle le groupe condensable est composé d'un ou plusieurs carboxy, carbalkoxy ou hydroxy.

13. Composition selon la revendication 1, dans laquelle le composé réactif est

14. Composition selon la revendication 1, dans laquelle le composé réactif est

$$(CH_3)_2N^{-1}$$
 $CO_2C_2H_5$ $CO_2C_2H_5$

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15. Composition selon la revendication 1, dans laquelle le composé réactif est

16. Composition seion la revendication 1, dans laquelle le composé réactif est

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$$(\mathsf{CH}_3 \overset{\mathsf{O}}{\mathsf{COCH}}_2 \mathsf{CH}_2)_2 \cdot \mathsf{N} \cdot \overset{\mathsf{CH}}{\overset{\mathsf{CH}}_3} \cdot \mathsf{CH} = \mathsf{C} \overset{\mathsf{CN}}{\mathsf{CO}}_2 \mathsf{CH}_3.$$

17. Composition selon la revendication 1, dans laquelle le composé réactif est

$$(CH_3COCH_2CH_2)_2-N-CH=CH_3$$

18. Composition selon la revendication 1, dans laquelle le composé réactif est

19. Composition selon l'uen quelconque des revendications 1 à 3 en mélange avec un agent de réticulation.

20. Composition réticulée selon la revendication 19.

21. Articles formés à partir de la composition réticulée de la revendication 20.